EFFECT OF CENTRIFUGAL FIELDS ON THE SELF DIFFUSION RATE OF SILVER

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INTRODUCTION

The results of research in the field of diffusion of solid materials, besides giving a better insight to the behavior of solid materials, lend themselves to many applications in the field of metallurgy. Several metallurgical processes which involve diffusion are: carburizing in which carbon diffuses into the steel, nitriding, manufacturing of bimetal strips which depend on adhesion resulting from interdiffusion of the two metals, and the rate of age hardening and transformation of metals. By relying more and more on purely scientific work, metallurgy is becoming less of an art and more of a science. It was therefore felt that some additional work toward the better understanding of the behavior of metals could provide not only a worthy contribution to physics but could also be used in the field of metallurgy.

The techniques developed by Dr. J.W. Beams of applying high centrifugal forces to thin films deposited on steel rotors seemed to be an ideal way to apply a directional force to metal samples without the usual twisting moments and force concentrations encountered in other methods. It was therefore decided to investigate the effects of a centrifugal field on the rate of diffusion of metals.

The two common methods used for measuring diffusion are the "direct" method and the 'indirect" method.

In the "direct' method two substances are placed in contact and maintained at a high temperature for a long period of time. The specimen is then sectionalized and the amount of the constituents is determined by chemical analysis.

In the "indirect" method a radioactive isotope of a substance is placed in contact with a second material and the concentration of the isotope in the second material is then determined by means of radioactive detection. This method was first employed by G. von Hevesy, F. Seith and A. Keil.[1] A lead isotope, Thorium B, emitting alpha rays, was used to determine the self diffusion of lead in a lead single crystal; noting the lessening of emissions as diffusion took place.

Three possible simple mechanisms for the diffusion of the atoms of one metal, A, through a metallic solid, B, are:

1) The A atoms may individually diffuse through the interstices of the solid, B;
2) The A atoms may exchange position with the B atoms within the lattice; and

3) The A atoms may move into adjacent vacant positions in the lattice of the solid, B.

In the case of the first mechanism the activation energy required to move an atom increases greatly with the smallness of the size of the interstitial sites. These sites are much smaller than the atomic size for metals forming substitutional alloys and therefore probably only metals forming interstitial alloys would lend themselves to this type of mechanism.

The activation energy required for the second mechanism would be of the same order of magnitude as the cohesive energy because the two atoms would be squeezed closely together by the surrounding atoms of the lattice during the exchange. The probability of this mechanism being responsible for diffusion is rather low considering the values of the activation energy obtained from numerous data, Seitz.[2]

The third mechanism for diffusion assumes that vacant sites, hereafter called vacancies, exist in the lattice of the material. This has been the topic of many investigations. Theoretical work in thermodynamics shows that the entropy, would be increased as a result of the increased disorder, introduced by the vacancies in the structure.

Although substitutional alloys are not expected to diffuse by the first mechanism, there is still the problem of determining which mechanism is responsible for their diffusion. Zener[3] has proposed a combination of mechanisms, in which more complex geometrical patterns of the motions of the atoms could be the means of diffusion.

Regardless of the mechanism involved, the relative displacement of the two kinds of atoms must be known to determine the rate of diffusion. A classical example of determining the rate of diffusion by measuring the relative displacement was accomplished by Smigelskas and Kirkendall.[4] They embodied molybdenum wires at the interface between copper and alpha brass. After maintaining the specimen at a high temperature for a long period of time, the molybdenum wires were found to have moved into the brass. This effect was observed for many other pairs of face center cubic metals. This is thought to be an indication that neither the direct exchange mechanism nor the Zener theory are the chief processes of diffusion in these metals.

In addition to the above described mechanisms, the effects upon the rate of diffusion of temperature and grain size must be considered.

A strong dependency of the rate of diffusion upon the temperature applies to both types of diffusion,

grain boundary and volumetric. This dependency is expressed by a relation of the form, $D = \text{Const.}\ e^{\frac{-q}{RT}}$; where R is the gas constant; T, the absolute temperature; and q, the activation energy which must be supplied before an atom may jump from one position to another in the lattice.

It was shown by W. A. Johnson[5] that the self diffusion constant for silver differed for grain boundary and volumetric diffusion. This is in agreement with the theory of Chalmers[6] based upon the behavior of very pure tin in the vicinity of its melting point. His results indicated that the melting point of the material in the grain boundaries was slightly lower than that of the bulk material. From these results it was concluded that the transition layer of atoms in the grain boundaries was thermodynamically less stable than the interior atoms of the grains. This would be in agreement with the previous remarks concerning increases of entropy with increased vacancy concentration.

THEORETICAL CONSIDERATIONS

The familiar laws of Fick for the concentration, C, of a solute in a solvent in terms of space time coordinates are:

$$J = -D\nabla C$$

and $$\frac{dC}{dt} = D\nabla^2 C$$

D is the diffusion constant which accounts for the grain size, type of diffusion occurring, and the lattice structure of the two media. It is a function of the temperature and the activation energy. J is the diffusion current.

The use of a collimating slit limits the peripheral length of the cylinder which is seen by the crystal. This allows the problems to be reduced to a one dimensional one and Fick's equations become:

$$J = -D\frac{dC}{dx}$$

$$\frac{dC}{dt} = D\frac{d^2C}{dx^2}$$

The following assumptions are made:

1) a unit line instantaneous source of radioactive silver at time zero and at $X = 1.76 \times 10^{-4}$ cm.;

2) no evaporation at the free surface, $X = 5.28 \times 10^{-4}$ cm.;

3) no diffusion through the boundary at the steel rotor; and

4) a uniform force field, ϕ, acts in the positive X direction.

By applying the above assumptions to Fick's one dimensional equations, the following are obtained:

$$J = -D \frac{\partial C}{\partial X} + C = 0 \text{ at } X = 0 \text{ for all } t$$

$$J = D \frac{\partial C}{\partial X} + C = 0 \text{ at } X = 5.28 \times 10^{-4} \text{cm. for all } t.$$

$$\text{and } \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial X^2} - \phi \frac{\partial C}{\partial X}$$

The solution of these equations is the same as that worked out by Carslow and Jeager[7] for the one dimensional heat conduction case. Applying the conditions of the present problem to their solution the following relationship of C is obtained:

$$C = \sum Z_n(x) \; Z_n(1.76 \times 10^{-4}) \exp. (-D \; t)$$

$$\text{where } Z_n(x) = \frac{2(D \cos x + \sin x)}{(5.28 \times 10^{-4}) D^2 + (5.28 \times 10^{-4})}$$

and the n's are the solution of the equation $\tan (5.28 \times 10^{-4}) = 0$ or $= \frac{n\pi}{5.28 \times 10^{-4}}$, $n = 1,2,3,4,.....$

To solve the equation numerically, values for ϕ and D have to be obtained. ϕ can be written as a constant times the force, where is a transport function

and F is the centrifugal force resulting from the rotation of the rotor. By substituting $= \frac{DN}{RT}$, (Jost[8]), and F equal to into $\phi \approx F$; ϕ equals $\frac{DN}{RT}\omega^2 rm$. This term becomes 6.1×10^{-10} D for T = 600 K. The space dependent equation reduces to:

$$Z_n(x) = 87.2 \quad \frac{\cos x \; \sqrt{6.1 \times 10^{-10}} \; \sin}{\sqrt{(6.1 \times 10^{-10})}}$$

The diffusion constant has been described by the equation:

$$D = D_0 \; e^{\frac{-U}{RT}}$$

where U is the activation energy; T, the absolute temperature; R, the universal gas constant; and D_0 a constant which varies very slowly with temperature and contains factors pertaining to different metals. The first and last quantities are found by fitting curves to experimental data.

Work has been done by several investigators to determine the self diffusion constants of silver by electrodepositing a radioactive silver layer on regular silver.

Hoffman and Turnbull[9] found that the diffusion constant for diffusion into high purity silver at 500° C with grain boundaries with a width of 5×10^{-8}cm. was:

$$D_{gb} = 0.03 \times e^{-\frac{20200}{RT}} \text{ cm}^2/\text{sec.}$$

Johnson[5] found the lattice self diffusion constant for silver to be

$$D_L = 0.895 \ e^{-\frac{45950}{RT}} \text{ cm}^2/\text{sec.};$$

and the combined diffusion for polycrystalline silver in a temperature range between 500 and 600 degrees centigrade to be

$$D = 2.3 \times 10^{-5} \ e^{-\frac{26400}{RT}} \text{ cm}^2/\text{sec.}$$

Using this last equation at room temperature, D equals 11.7×10^{-20} cm^2/day which compares favorably with D equal to 9.6×10^{-20} cm^2/day noted by Harrier.[10]

EXPERIMENTAL TECHNIQUE

The method of using a radioactive isotope was decided upon to detect the self diffusion of silver. The main advantage of this method over sectionalization or chemical analysis was to allow the determination of the rate of diffusion at numerous time intervals during each run without damaging the film. Two major problems had to be considered: the method to be used for depositing the films; and the method of obtaining data to be correlated with the rate of diffusion.

Two possibilities presented themselves for depositing the films: electrodeposition and evaporation.

An evaporation technique would have required a large bell jar containing not only the test rotor and the evaporation equipment but also a mechanism for rotating the rotor during the evaporation to insure even thicknesses of the deposited layers. Although this technique is readily adaptable for non-radioactive materials, it is not practical for use with radioactive materials because the amounts of the radioactive isotope deposited on the surface of the bell jar and mechanisms results in a wasteful usage of the isotope, and even more important presents an excessive radiation hazard to the worker while cleaning,

decontaminating, and disposing of the radioactive wastes.

Because electrodeposition would present much less of a radiation hazard and would be much more economical at the same time, it was decided to investigate this process for obtaining the desired films.

In the early trials of electroplated rotors the films were thrown from the rotors at various speeds. The initial procedure adopted for the electrodeposition of films on the rotors was to:

1) polish the rotor with single ought metallurgical polishing paper;

2) wash the rotor in acetone and rinse with distilled water;

3) wash the rotor in hot detergent and rinse with distilled water;

4) deposit a thin layer of copper by chemical deposition for a period of three seconds in an Acid Copper Plating Solution,[11] (This solution was prepared by diluting 3.5 grams of copper sulphate crystals and 8.0 ml. of concentrated sulphuric acid to a final volume of 100 ml.);

5) electroplate an initial non-radioactive silver film from a silver strike solution,[12]

(This solution was prepared by dissolving 509 milligrams of silver cyanide and 37.5 grams of sodium cyanide in 500 ml. of distilled water.);

6) electroplate the bottom and cylindrical surface of the rotor with a film of radioactive silver, ordinary silver enriched with silver^{110} from a silver cyanide plating solution prepared by dissolving 30.4 grams of radioactive silver cyanide, 41.1 grams of potassium cyanide, and 44.8 grams of potassium carbonate per liter of solution;

7) and electroplate the final coating of non-radioactive silver over the entire rotor from the plating solution described in 5) above.

The thickness of the films was determined from the following equation which was derived from Faraday's Law;

$$T = \frac{M\,a}{F\,\rho\,A}\,t$$

where T is the thickness of the film in cm.; M, the molecular weight of silver; a, the current in milliamperes; F, the Faraday constant; ρ, the density of silver; A, the area to be plated; and t, the plating time in seconds. The current density was maintained constant at 8.62 ma/cm^2 for this experiment and the plating times were varied to

give the various layer thicknesses. The thickness of the initial layer was 6.92×10^{-5} in.; the radioactive layer, 4.58×10^{-6} in.; and outer layer, or overlay, 1.38×10^{-4} in. Microscopic examination revealed that the films prepared from these solutions were polycrystalline with a grain size less than one micron.

The relative movement of the radioactive isotope through the ordinary silver layers was determined by counting the -ray emission with a scintillation crystal, energy discriminator and counting device, described under "Description of Apparatus." A slight and very broad peak appeared at the same place in the energy spectrum of the emissions from several samples. By incorporating a collimating slit and a very thin aluminum reflector, this peak was narrowed considerably. A typical energy spectrum obtained with the collimating slit is shown in Figure 1.

It was found that various thicknesses of the overlay lowered the counting rate at each point on the energy curve but did not produce any detectable shift of the peak of the curve towards higher or lower energies. A set of these curves is shown in Figure 2. From these results, the parameter chosen for the determination was the measurement of the number of counts in a wide energy band chosen, so as to include the peak of the energy curve. This energy

band ranged between two points where the number of counts were the same and the slopes were approximately equal and of opposite sign. This compensated for small shifts of the energy spectrum caused by voltage and heat fluctuations in the counting equipment. Refering to Figure 1, this band ranged from 10.0 to 17.5 volts (Pulse-Height Analyzer settings).

To obtain the relation between the counting rate and thickness of overlay, eight rotors were plated with the radioactive isotope. The energy spectrum and a total count of the above defined energy band of each were taken. The rotors were then plated with overlays of various thicknesses and the spectra and total counts of the band were again taken. The peaks of the spectra, both before and after the overlay, of all rotors were found to lie at the same energy position. One of the rotors was chosen as a standard and the ratios of the initial number of counts of the other rotors to the initial number of counts of this standard rotor were subsequently used to compensate for differences in the amount of radioactive silver deposited on each rotor. The results of this investigation showed that a curve of the logarithm of the number of counts versus the thickness of the overlay turned out to be a straight line, as illustrated in Figure 3.

A control rotor was prepared by depositing a layer of radioactive silver on it. This was maintained at room temperature and counted each time a test rotor was counted. The actual number of counts of the test rotor were then adjusted by the ratio of the actual number of counts of the control rotor at this time to its initial value at the beginning of the run. This compensated for the decrease of the counting rate with time resulting from radioactive decay in the test films during long runs. This procedure further compensated for drift in the counting equipment that may have taken place during the run.

Thus the following procedure for detecting a shift in the position of the radioactive isotope was adopted. The films were plated on the test rotor, the number of counts in the energy band were totaled, and the number of counts in the energy band of the control rotor were totaled. Then the test rotor was placed in the centrifuge and brought to the desired speed and temperature. At appropriate time intervals the rotor was stopped, removed from the centrifuge, its energy band count taken and a parallel count of the control rotor was taken. The test rotor was then replaced in the centrifuge, and returned to

the desired speed and temperature. After adjusting the number of counts of the test rotor, as mentioned above, the percent change in counting rate of emissions from the test rotor with respect to its counting rate at the beginning of the run was computed.

A run was conducted as follows with the rotational velocity of the spinning portions at 10,000 revolutions per second. The rotor was counted at four hour intervals throughout this run. For the first twenty-four hours, with the rotor spinning, the counting rate decreased. Since it was expected that the counting rate would increase, the centrifugal force increasing the rate of diffusion towards the periphery, instead of decreasing, the rotor was suspended in the centrifuge at the same temperature without spinning for a period of eight hours. A rise in the counting rate was noted. The rotor was then spun for a period of eight hours and a decrease in the counting rate was noted. Then the rotor was suspended, without spinning, for a period of twenty hours and the counting rate increased again. This is illustrated in Figure 4. A parallel run under similar conditions produced the same effects.

Several runs were made with the rotor suspended at temperature without spinning, for seventy-two hours and the counting rate exhibited a slight increase throughout

the runs. Since there was a change from an increasing counting rate at zero rotational velocity to a decreasing counting rate at 10,000 revolutions per second, the change in counting rate with time for rotational velocities of 2,000; 4,000; 6,000; and 8,000 revolutions per second was investigated. These runs lasted for periods of seventy-two hours, with the rotor being counted at twelve hour intervals. The results of these runs are shown in Figure 5.

In order to determine whether any of these effects were the result of plastic flow of the material plated on the cone and the bottom of the rotor, a method was developed for plating only the cylindrical surface of the rotor. This was accomplished by machining a drilled holder of soft steel which fitted the cone of the rotor, and a plate undercut and fitted against the bottom of the rotor. The two items were of the same outside radius as that of the rotor and were coated with polystyrene to prevent silver from plating on them. The three parts of the assembly were held together by the magnet which also held the assembly to the stirring motor in the plating bath.

In addition to limiting the silver plating to the cylindrical surface of the rotor, this method of plating

also produced a more uniform layer of deposited silver in that no sharp corners were now present and therefore the edges of the cylinder were no longer points of high current density.

It was discovered at this point that the copper plate could be omitted and hereafter it was not used.

The results of another set of runs made at 0; 2,000; 4,000; 6,000; and 8,000 revolutions per second using this latter plating technique are shown in Figure 6.

DESCRIPTION OF APPARATUS

The apparatus used in this experiment can be subdivided into three groups:

1) the plating apparatus for electrodepositing the silver layers on the rotors,

2) the ultracentrifuge used to apply large centrifugal forces on the test rotors, and

3) the apparatus used to detect and count the radioactive emissions from the test rotors.

Plating Apparatus

A water bath kept at constant temperature by a mercury thermoswitch controlled nichrome heating element maintained the plating solution at the proper plating temperature. The framework holding the plating solution beaker in the temperature bath was designed to allow rapid removal of the beaker and insertion of the radioactive plating solution container. A six revolution per minute motor was provided to rotate the test rotor in the plating solution to insure uniform thicknesses of the silver films.

The radioactive plating solution was enclosed in a thick brass container to lessen the amount of radiation exposure to personnel. A platinum anode was employed to

reduce to a minimum the variations in the ratio of radioactive silver to non-radioactive silver in the solution. During storage, a tight fitting brass cap over the solution container kept the evaporation of the solution to a minimum. The circuitry of the plating apparatus is shown in Figure 7.

The Centrifuge

This experiment made use of the magnetic suspension, magnetic driven ultra-centrifuge constructed by O.R. Harris.[13] Figure 8 shows an overall view of the centrifuge and related equipment.

The support system consists of a large solenoid mounted above the rotor and, mounted beneath the rotor, a coil in the grid tank of a tuned-grid-tuned-plate oscillator whose output controls the current flowing through the solenoid. As the rotor changes vertical position, the induction in the grid tank coil is varied, detuning the oscillator. This results in an oscillator output signal which is a function of rotor height. A derivative circuit was employed to minimize overshoot and to increase vertical stability. The bias on the derivative amplifier, V-103, was increased from 1.5 volts DC to 4.5

volts DC. This increased the stability to such an extent that the centrifuge could be left operating unattended for periods of twelve hours. The support circuit and support power supply circuit are shown in Figures 9 and 10.

Horizontal damping was accomplished by suspending a steel core in the center of the support solenoid by means of fine hypodermic tubing. The end of the core was immersed in oil to provide a means of absorbing the energy of the horizontal oscillations.

The drive circuit consisted of a phase shift oscillator feeding a signal into a phase splitting network. Three signals, each one hundred twenty degrees out of phase with each other, were then amplified and then applied to three pairs of drive coils. The eddy currents induced in the rotor caused the rotor to accelerate and try to catch up with the rotating field. The amount of acceleration was controlled by varying the amplitude of the output sine wave of the oscillator which oscillated at a fixed frequency of 35.4 kilocycles. The circuit diagram for the oscillator is shown in Figure 11. The phase splitting circuit was changed to provide separate control for the cathodes of the power amplifiers.

Better control of the inputs to the separate drive coils pairs was obtained, providing a better adjustment for balancing the drive field. A conventional 300 volt DC regulated power supply with an output of 400 milliamperes at one percent regulation was used. The phase splitting and drive power supply circuits are shown in Figures 12 and 13.

The evacuated chamber, situated between the six drive coils, contained a tungsten coil used to raise and maintain the temperature of the rotor by radiation heating. A method of mounting the tungsten heating coil in the test chamber was devised in order that the heating coil would be able to absorb shock if it were struck by a spinning rotor. This prevented breakage of the spot-welded joints of the tungsten leads.

The temperature of the rotor was determined by using a Rubicon potentiometer to measure the temperature of a similar rotor mounted on an iron-constantan thermocouple within the vacuum system. This rotor was heated by a similar tungsten coil connected in series with the one in the test chamber. This temperature was corrected for heat conduction along the thermocouple and for radiation from the coils surrounding the test chamber.

A light beam from a D.C. source entered the chamber through the center of one of the drive coils, was reflected off the rotor through the center of another drive coil to a phototube. The amplitude pattern from the phototube was beat against a known pulse on an oscilloscope to determine the rotational velocity of the rotor.

A conventional vacuum system consisting of a mechanical fore pump, dryer containing phosphorus pentoxide crystals, oil diffusion pump, fore pressure thermocouple gauge and a high vacuum ionization gauge was used. Vacuums of the order of 1×10^{-6} mm of mercury were obtained. The system also incorporated a high vacuum stop cock, ground glass joint, and an air predrying inlet system which allowed removal and replacement of the rotor with a minimum loss of time for the pumps to obtain the operating vacuum level.

A diagram containing the dimensions of the test rotor is shown in Figure 14. It consisted of a truncated cone machined on tops of a cylinder. Proper adjustment of the equipment proved that this configuration had very stable support characteristics in the magnetic field used for suspension. Two such rotors were machined from a rod of tool steel, each being attached to a short mandrel by a tapered sprue. This facilitated polishing the cylinder

and cone. The bases were polished after breaking the rotors loose from the mandrel and mounting them in a drill press by means of a Teflon collet. There was no need for heat treating the rotors as the velocities and temperatures at which the experiment was performed were well below the bursting speed of the rotors.

Beta-Ray Counting Apparatus

The beta-ray counting apparatus, shown in Figure 15 and schematically in Figure 16, consisted of the following:

1) light tight compartment housing the rotor, stilbene scintillation crystal and photomultiplier tube;

2) linear amplifier;

3) pulse height analyzer;

4) scaler;

5) timer.

The rotor was held in position within the light tight compartment on a spring loaded plate which held the rotor cone in the cap and shielded the radiation from the base of the cylinder. See Figure 17.

After much experimenting it was found to be almost impossible to position the rotor so that the identical area was presented to the crystal each time the emissions were counted. Both the bottom plate and conical cap shafts were mounted in bearings and a one revolution per minute motor was connected to the conical cap shaft to rotate the rotor. This motor operated automatically when the "count" switch on the scaler was engaged. Counting periods lasted for two minutes which allowed the rotor to be turned through two complete revolutions. A one-quarter inch thick lead shield, mounted between the rotor and the stilbene crystal and containing a collimating slit with dimensions of the height of the rotor cylinder and width of one tenth the rotor diameter, limited the area of the cylinder seen by the scintillation crystal.

A thin stilbene crystal was selected for the scintillation crystal because of its relative insensitiveness to gamma radiation. A thickness of one-half millimeter was selected to provide an escape peak for the 0.53 MEV electrons which have a range of 0.86 millimeters in stilbene. A twenty-five hundred thousandth of an inch aluminum foil mounted between the lead shield and the crystal served both

to absorb 0.030 MEV of the energy of the beta-rays[14] striking the crystal and to reflect the photons emitted in the crystal back to the photomultiplier tube. The output of the regulated negative power supply shown in Figure 18 was lowered from -1850 volts D.C. to -960 volts D.C. by lowering the potential difference between adjacent dynodes to eighty volts. By operating the DuMont 6291 photomultiplier tube at this lower value rather than near the maximum 2100 volts D.C., a very low noise to signal ratio resulted and the linear amplifier was adjusted to give maximum amplification without being overdriven. A cathode follower was used to match the high impedance of the photomultiplier tube to the low capacity coaxial cable carrying the signals to the linear amplifier.

The regulated negative power supply was rebuilt to incorporate advantages found by Dr. C. R. Harris doing experimental work at Electronics Research, University of Virginia. This consisted of replacing the original resistor voltage divider network by one consisting of resistors and NE 2 voltage regulator tubes to provide a more constant potential difference between the dynodes of the photomultiplier tube while the load is changing. See Figure 19. This resulted in a still lower noise to signal ratio while

the potential difference between dynodes was raised to 120 volts. The amplification required from the linear amplifier was thereby decreased. This seemed to lower the drift of the equipment and the data was more consistent.

A linear amplifier was constructed according to the schematic diagram Figure 20. The amplifier section of the Atomic Instrument Company's Model 240-C Linear Amplifier was utilized in conjunction with a medium voltage stabilized power supply,[15] having an output of plus 300 volts D.C. and 150 milliamperes available for plate currents. This proved to be a very stable combination with low rms ripple voltage. The bottom of the chassis was enclosed and a blower, rated at 50 cubic feet of air per minute, was mounted on the chassis. This forced air over a two hundred watt heating element past internal baffles through the amplifier section. A thermostat, cutting on at 109 degrees and off at 110 degrees Fahrenheit mounted in the air-stream under the amplifier section and connected in series with the heating element, provided a more even temperature of the amplifier components, thereby stabilizing the amplifier gain.

An Atomic Instrument Company Model 510 Pulse Height Analyzer was used to select the energy range desired.

The number of pulses received from the analyzer was registered on an Atomic Instrument Company Model 1030A Binary Scaler.

The time interval for the counting period was determined by noting the number of seconds and tenths of seconds registered on a Precision Scientific Company "Time-It" timer. The timer operated whenever the "count" switch on the scaler was engaged.

DISCUSSION OF RESULTS

A graphical representation of the results is shown in Figure 21, where the distance moved of the average radioactive particle is plotted against the applied force in dynes. The time is constant along each curve.

The distance moved by an average radioactive particle was computed by applying the change in counting rate to the curve of Figure 3. A sample calculation is shown in Appendix II.

As the force increases, the average distance moved increases to a maximum and then decreases. The maximum average distance moved occurred between applied forces of from 14 x 10^7 to 22 x 10^7 dynes. This is the range of forces at which the films with no adhesion were thrown from the rotor. This is also in agreement with tensile strength experiments performed in this laboratory by W. L. Hart.

The maxima of the curves obtained from the rotors plated by the first method were in all cases higher than those obtained from rotors plated by the second method.

Results such as the above are not evidently justifiable from the consideration of the time-independent part of the solution for the concentration of the diffusing medium. It is shown in the sample calculations that at a speed of 6,000 revolutions per second, the effect of the applied centrifugal force would alter this solution in the order of 10^{-4}, making the direct effect of the force negligible.

The time-dependent part of the solution of the concentration of the diffusing medium, contains the diffusion constant as a parameter. As has been previously pointed out, the diffusion constant depends upon the grain structure. Therefore, the above effects would indicate that the diffusion constant varied with the applied force. It is believed that cold work and recrystallization resulting from the stresses introduced by the applied centrifugal force causes rearrangement in the size and position of the grain boundaries. This could decrease the rate of diffusion.

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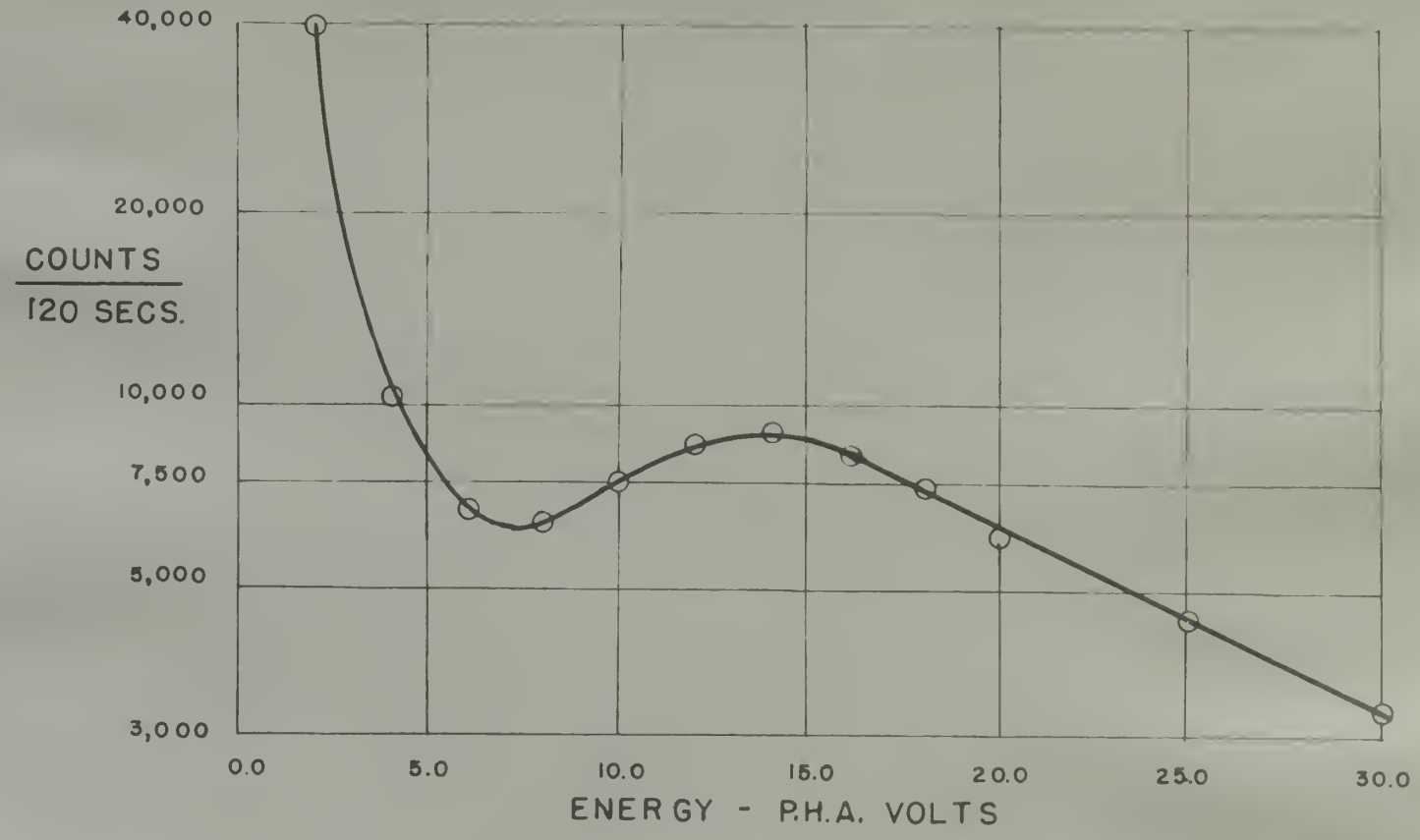
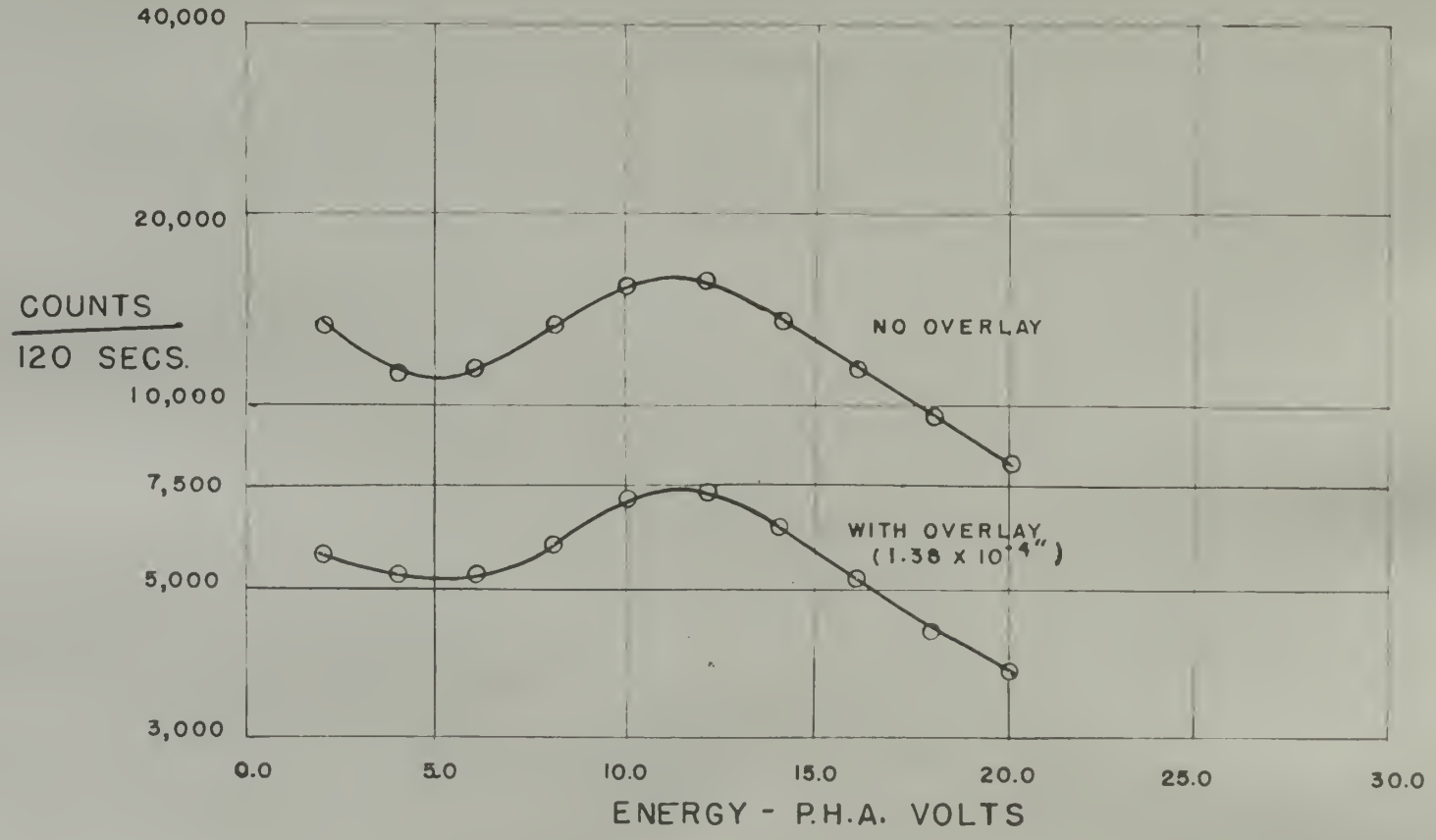
16. [illegible]



TYPICAL ENERGY SPECTRUM OF A PLATED ROTOR

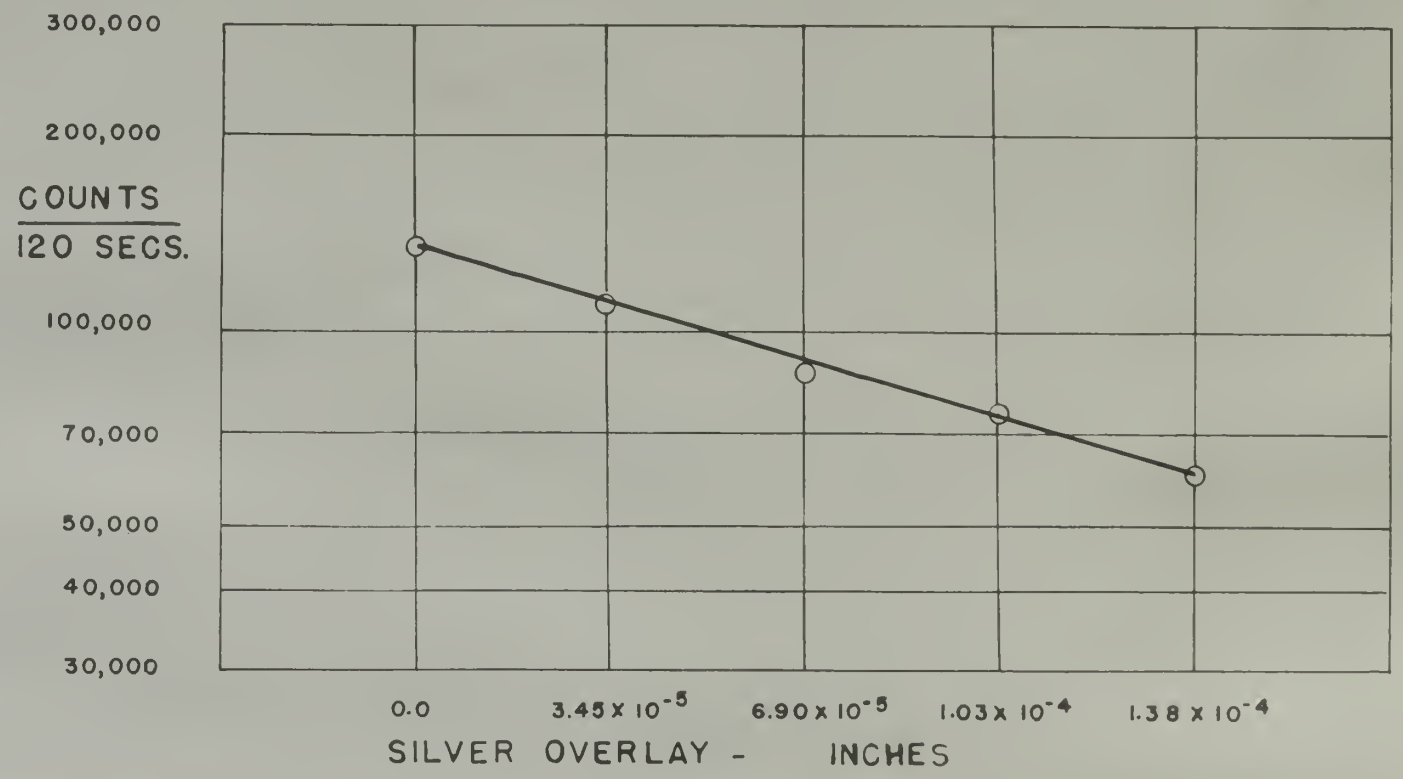
FIGURE 1

JJC



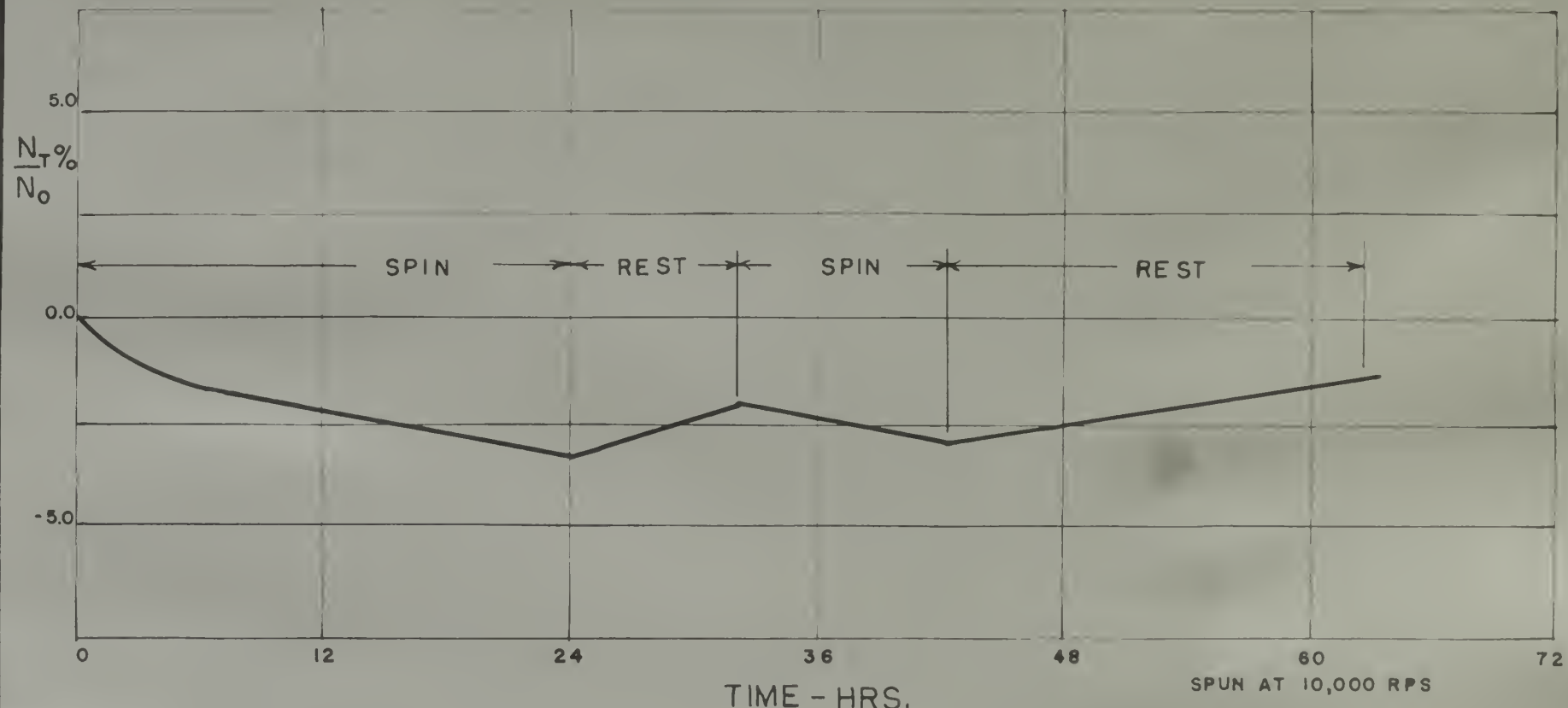
EFFECT OF OVERLAY OF NON-RADIOACTIVE SILVER

FIGURE 2



ENERGY ABSORPTION BY SILVER OVERLAY

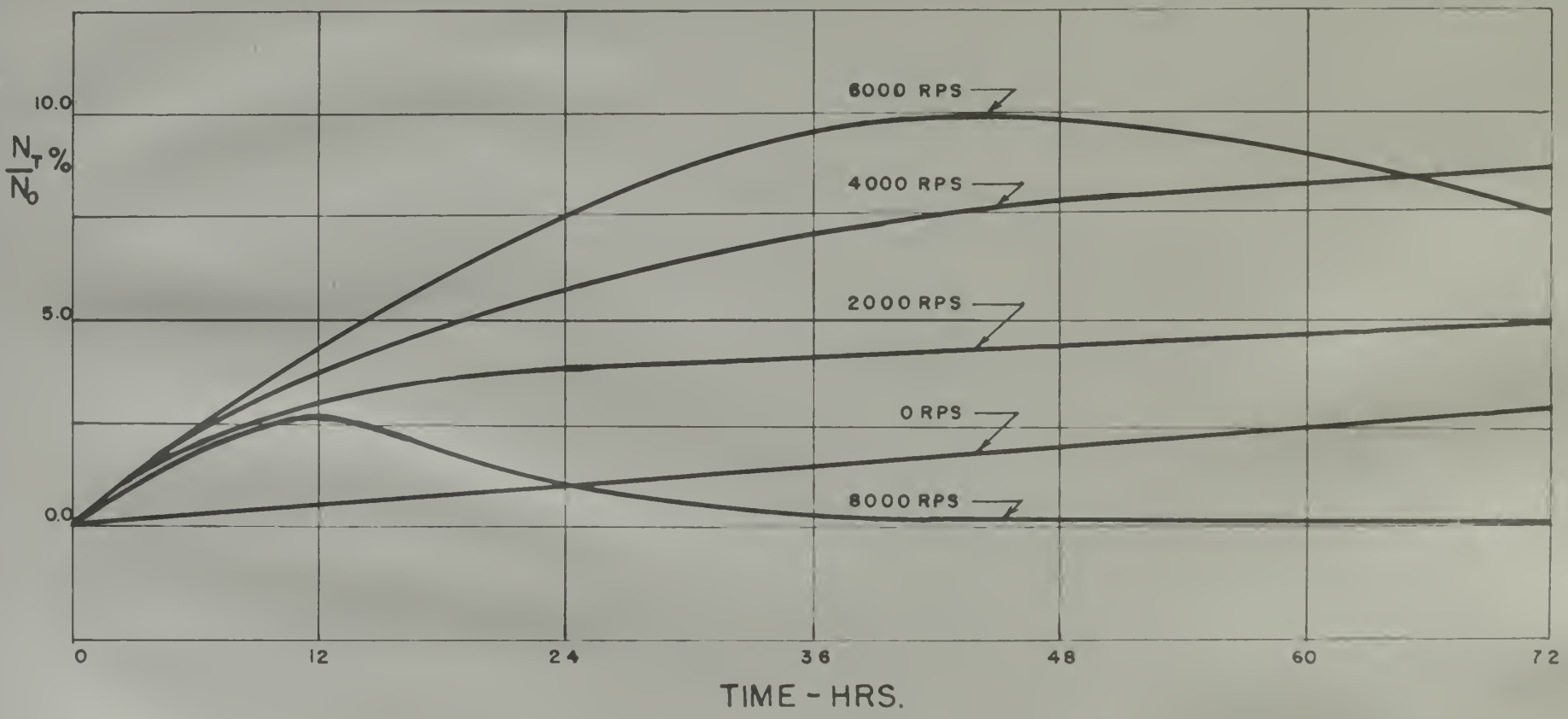
FIGURE 3



AG THROUGH AG
TEMP. 300° C

PERCENT CHANGE IN COUNTING RATE VS. TIME

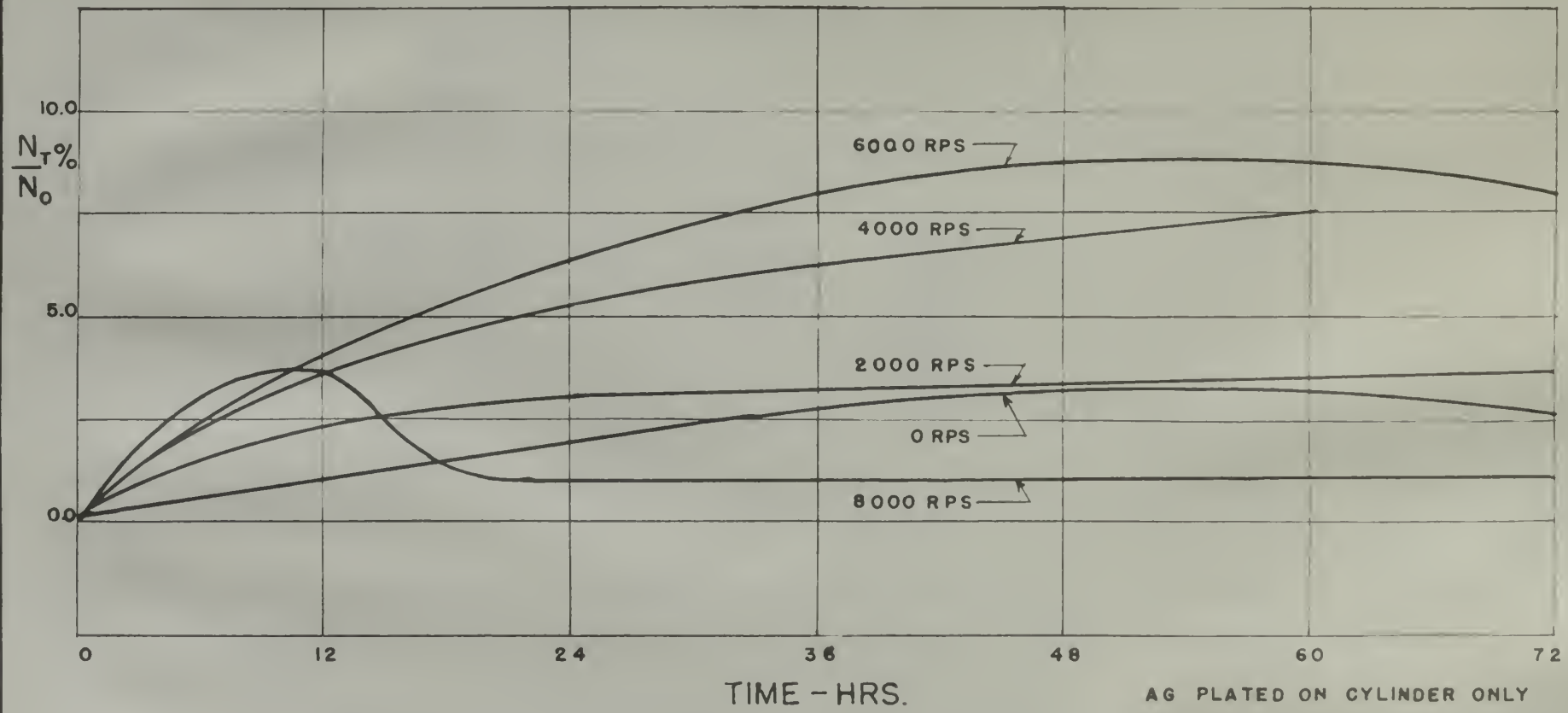
FIGURE 4



AG^{110} THROUGH AG
TEMP. 300° C

PERCENT CHANGE IN COUNTING RATE VS. TIME

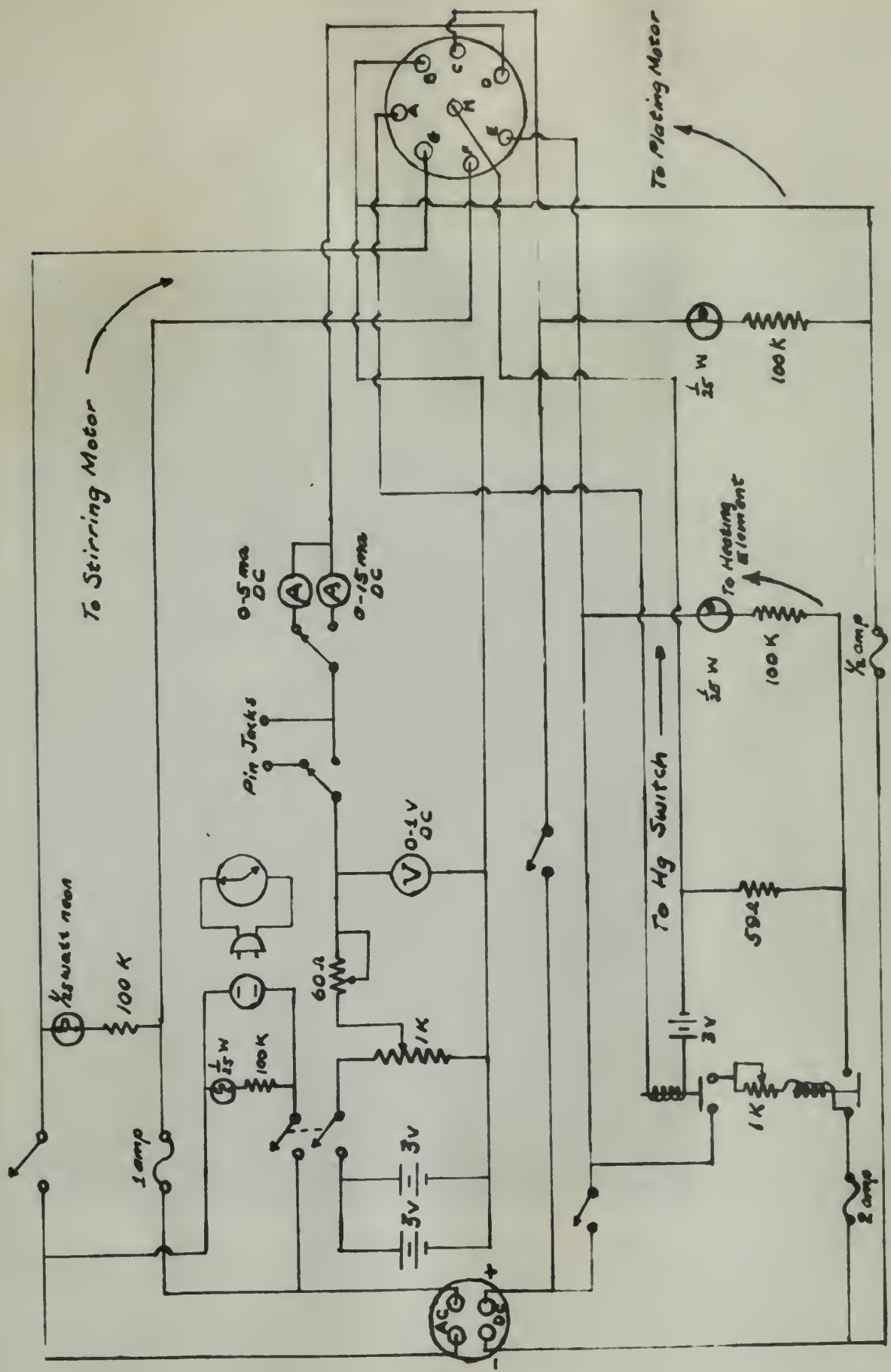
FIGURE 5



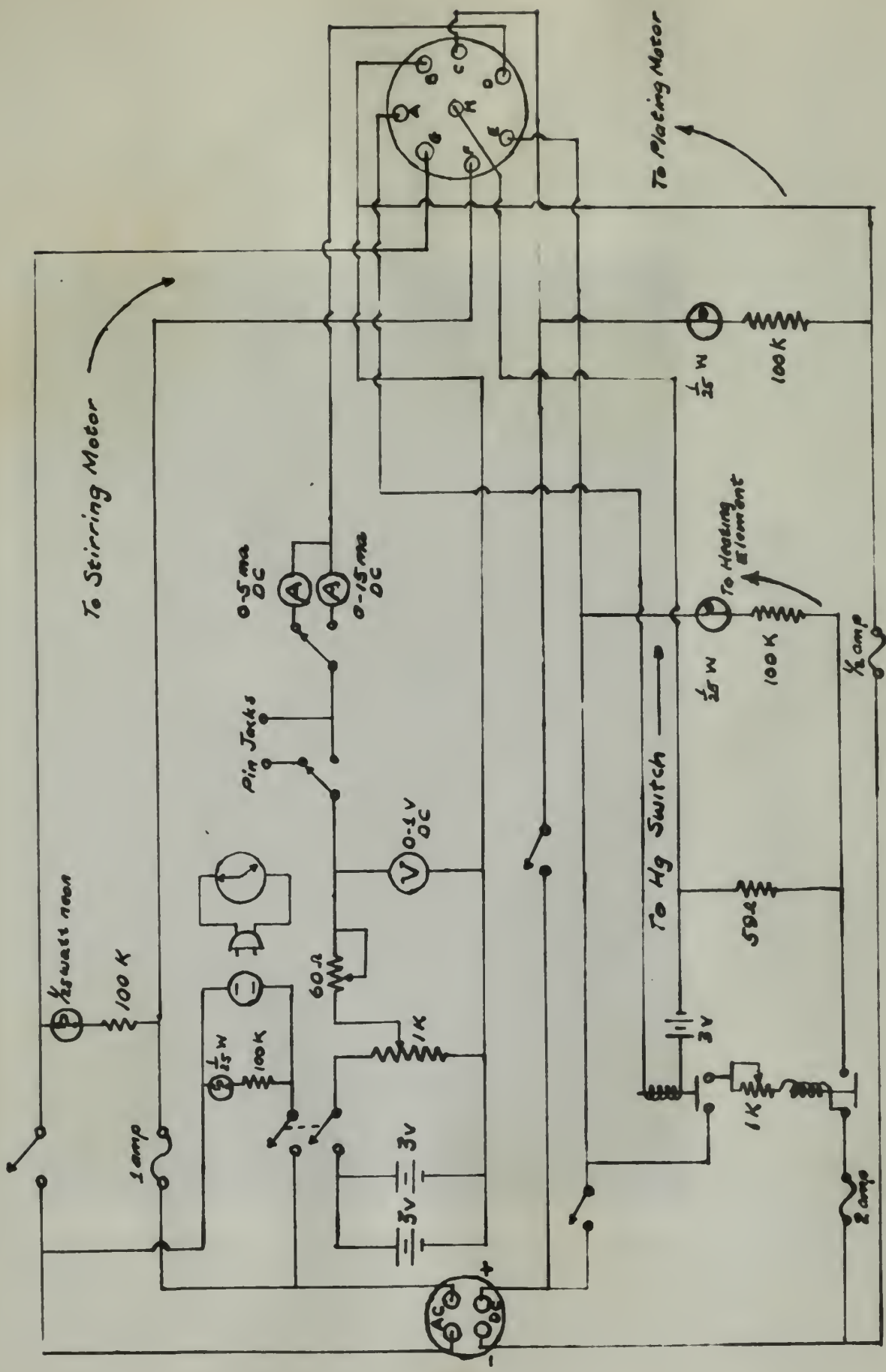
AG^{110} THROUGH AG
TEMP. 300° C

PERCENT CHANGE IN COUNTING RATE VS. TIME

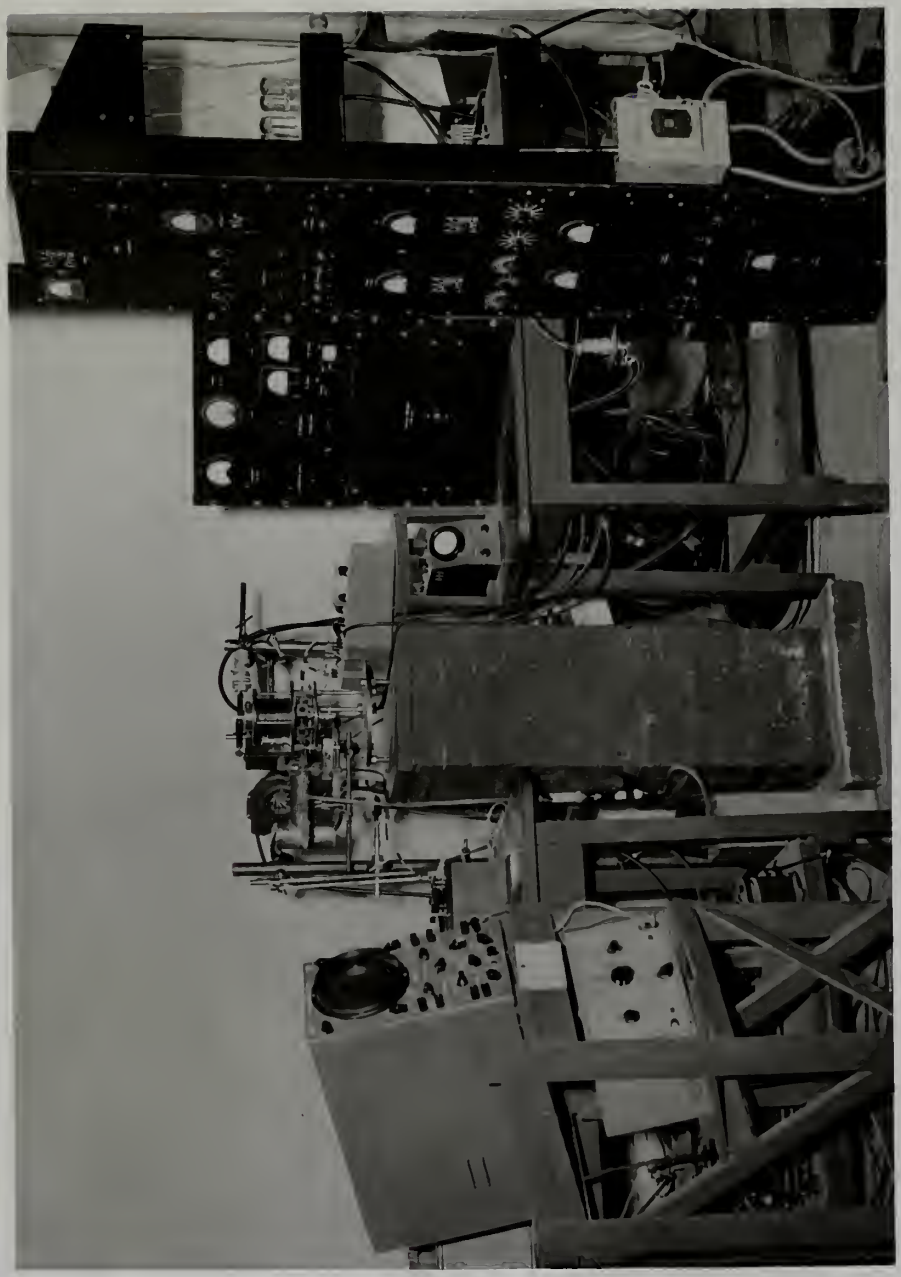
FIGURE 6



PLATING CIRCUIT



PLATING CIRCUIT



THE ULTRACENTRIFUGE - FIGURE 8

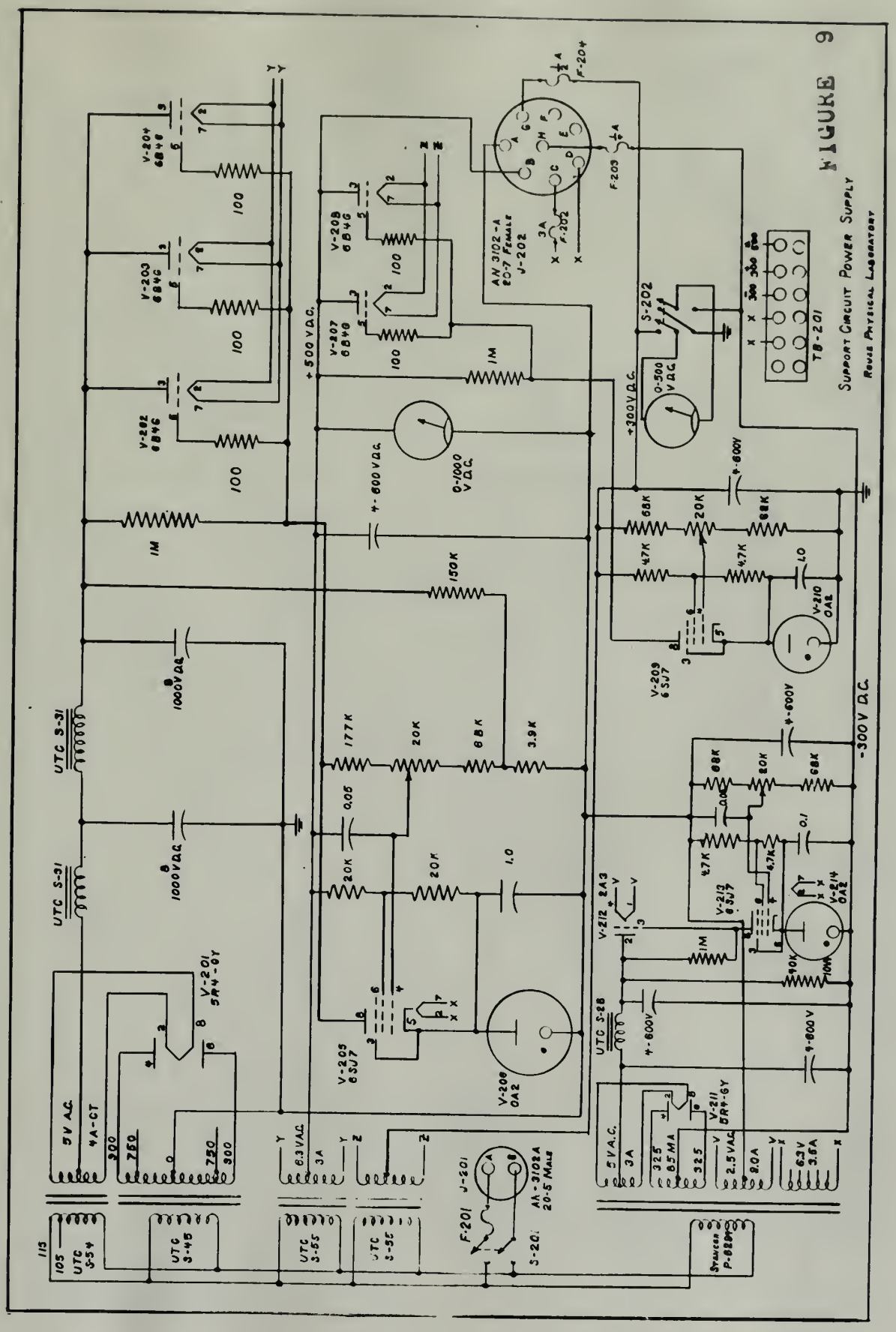


FIGURE 9
SUPPORT CIRCUIT POWER SUPPLY
ROYCE PHYSICAL LABORATORY
-300 V D.C.
+500 V.D.C.
+300 V.D.C.
+ 600 V.D.C.
0-1000 V.D.C.
0-500 V.D.C.
1000 V.A.C.
1000 V.A.C.
UTC S-31
UTC S-31
5V A.C.
4A-CT
6.3 V.A.C.
3A
5 V.A.C.
3A
325
65 MA
325
2.5 V.A.C.
8.0A
6.3V
3.6A
V-201 5R4-GY
V-200 0A2
V-205 6SJ7
V-206 6SJ7
V-207 6BQ6
V-208 6BQ6
V-202 6BQ6
V-203 6BQ6
V-204 6BQ6
V-209 6SJ7
V-210 0A2
V-211 BR4-GY
V-212 2A3
V-213 6SJ7
V-214 0A2
AN 3102-A 20-7 FEMALE J-202
AN-3102A 20-5 MALE J-201
S-202
S-201
F-201
F-203
F-204
TB-201
UTC S-28
UTC S-45
UTC S-55
UTC S-55
SPRAGUE P-88P8
115
105
UTC S-54
177K
20K
68K
3.9K
150K
1M
100
68K
20K
68K
47K
4.7K
10K
40K
0.05
0.1
1.0
4+600V
9+600V

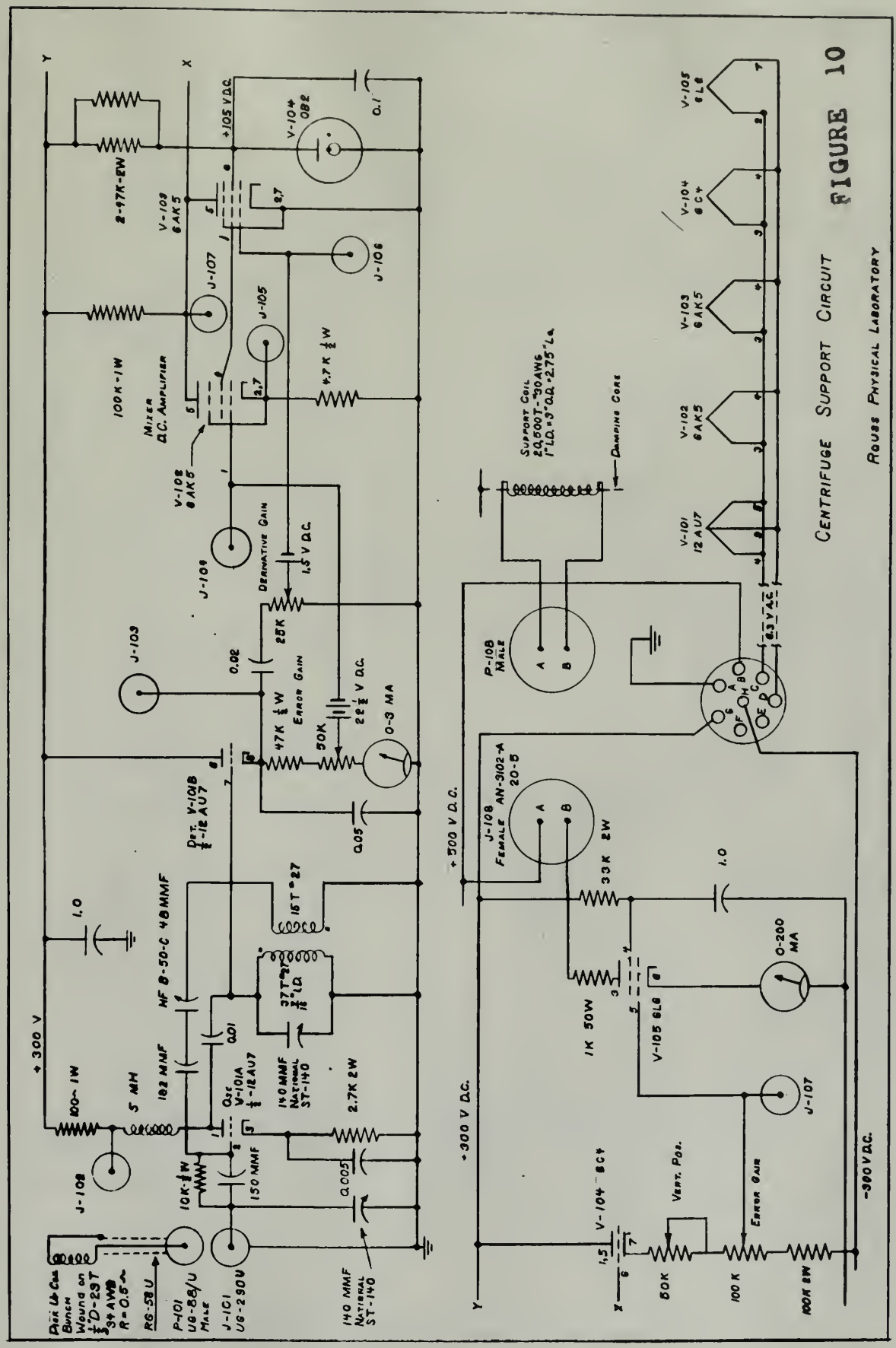

CENTRIFUGE SUPPORT CIRCUIT
ROUSS PHYSICAL LABORATORY
+300 V
+300 V D.C.
-300 V D.C.
+105 V D.C.
63 V A.C.
1.5 V D.C.
22½ V D.C.
0-3 MA
0-200 MA
2-17K-2W
100K-1W
47K ½W
25K
50K
1K 50W
33K 2W
2.7K 2W
10K-½W
182 MMF
150 MMF
140 MMF
5 MH
0.01
0.02
0.05
0.005
0.1
1.0
4.7 K ½W
2.7
Derivative Gain
Error Gain
Vert. Pos.
Mixer D.C. Amplifier
Det. V-108 ½-12AU7
Osc. V-101A ½-12AU7
HF 8-50-C 48MMF
140 MMF National ST-140
Support Coil 20,500T-30AWG 1"I.D.x3"O.D.x2.75"L.
Damping Core
J-101 UG-88/U Male
J-108 Female AN-3102-A 20-5
P-108 Male
RG-58U
V-101 12AU7
V-102 6AK5
V-103 6AK5
V-104 6C4
V-105 6L6
V-106 6AK5
V-107 6AK5
V-108 6AK5
V-109 0B2
J-101
J-102
J-103
J-104
J-105
J-106
J-107
J-108
X
Y

FIGURE 10

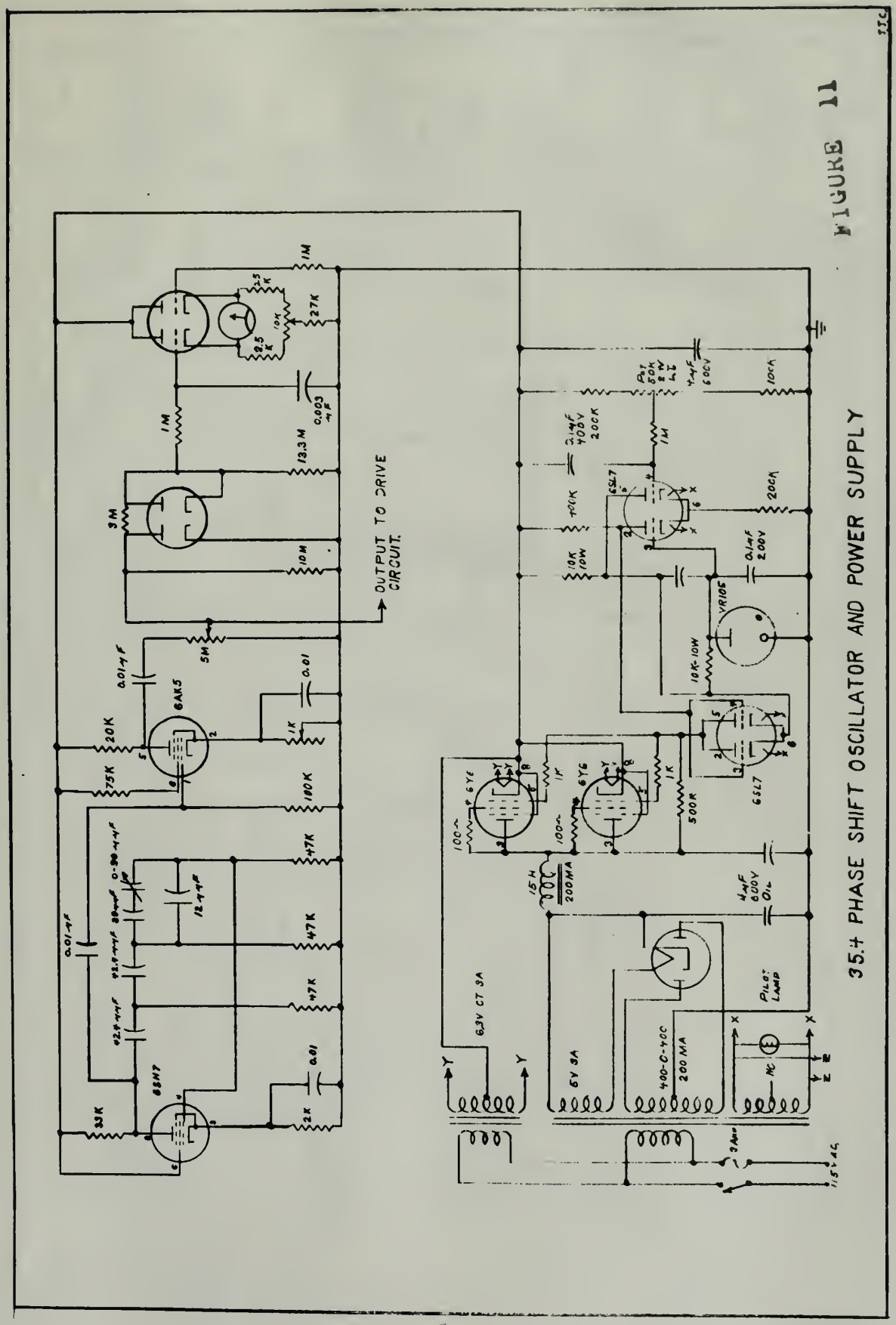
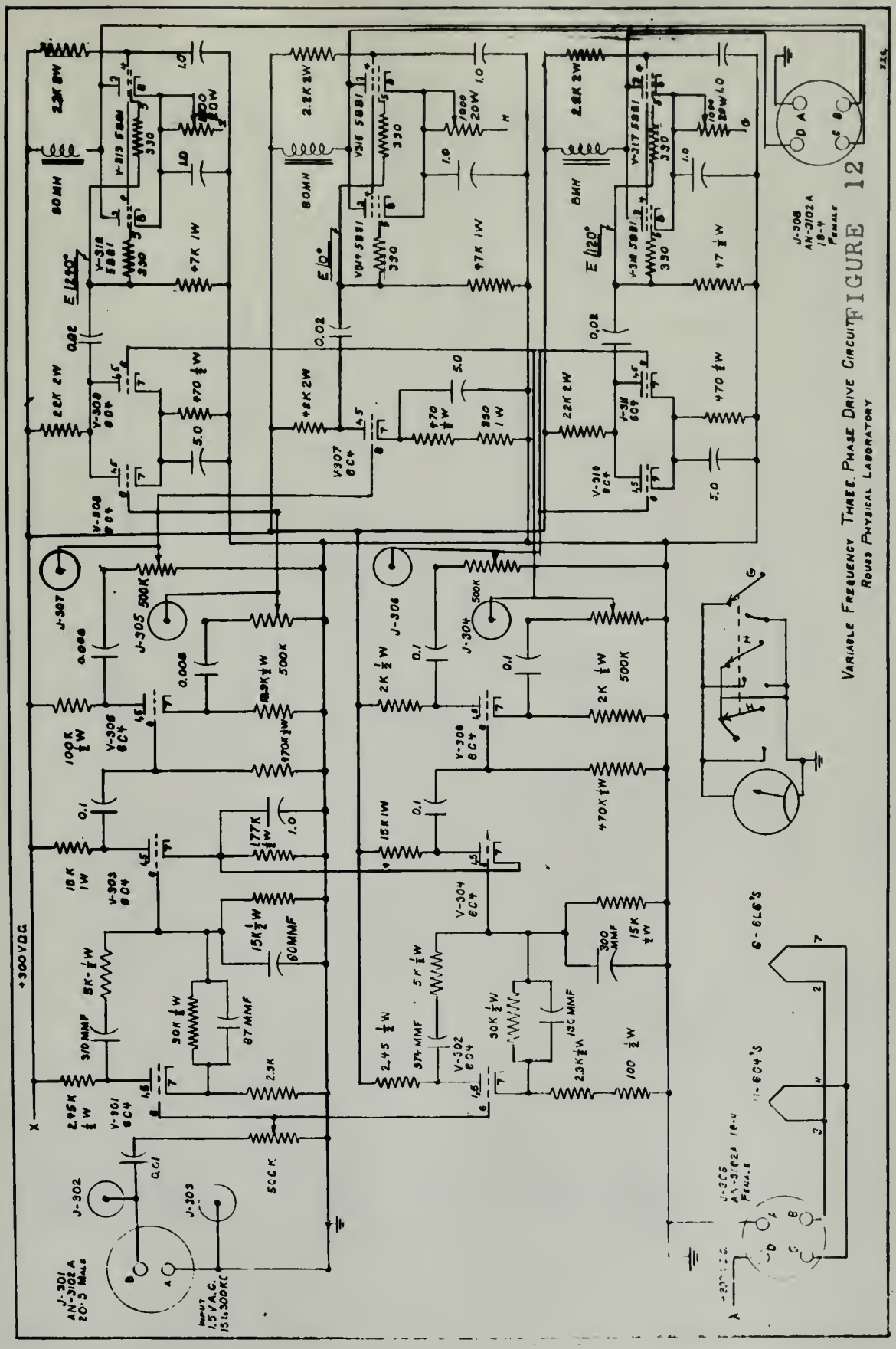


FIGURE 11

35.4 PHASE SHIFT OSCILLATOR AND POWER SUPPLY



VARIABLE FREQUENCY THREE PHASE DRIVE CIRCUIT FIGURE 12
ROUSS PHYSICAL LABORATORY

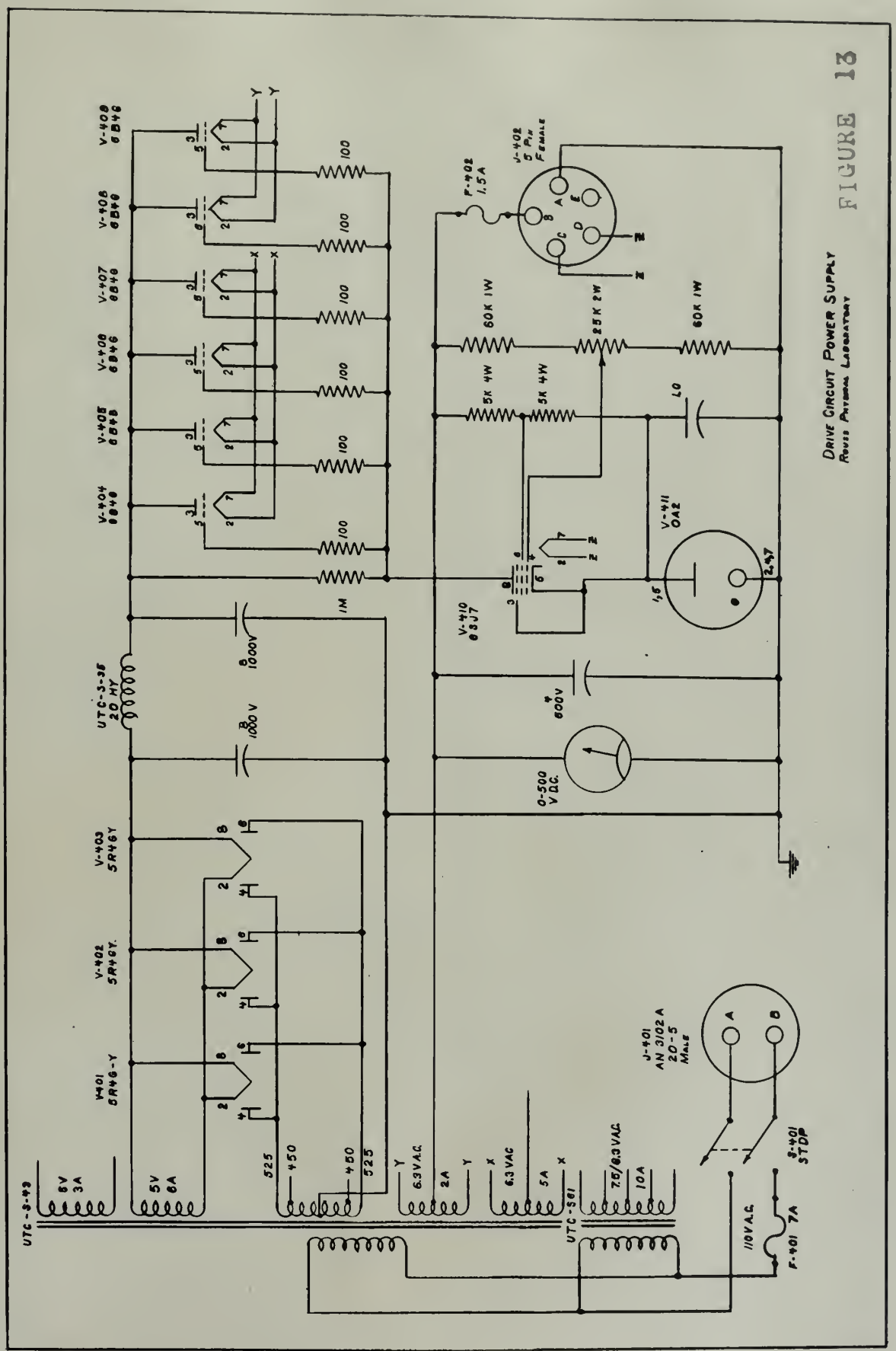

DRIVE CIRCUIT POWER SUPPLY
ROCKET PROPULSION LABORATORY
UTC-S-93
UTC-S-61
UTC-S-35
20 HY
V-401 5R4G-Y
V-402 5R4GY
V-403 5R4GY
V-404 6B4G
V-405 6B4G
V-406 6B4G
V-407 6B4G
V-408 6B4G
V-410 6BJ7
V-411 0A2
F-401 7A
F-402 1.5A
J-401 AN 3102A 20-5 MALE
J-402 5 PIN FEMALE
S-401 STOP
110 V.A.C.
6V 3A
5V 6A
525
450
6.3 VAC
2A
5A
7.5/6.3 VAC
10A
100
1M
60K 1W
25K 2W
5K 4W
0-500 V D.C.
600V
1000V
2.7
1.5
A
B
C
D
E
F

FIGURE 13

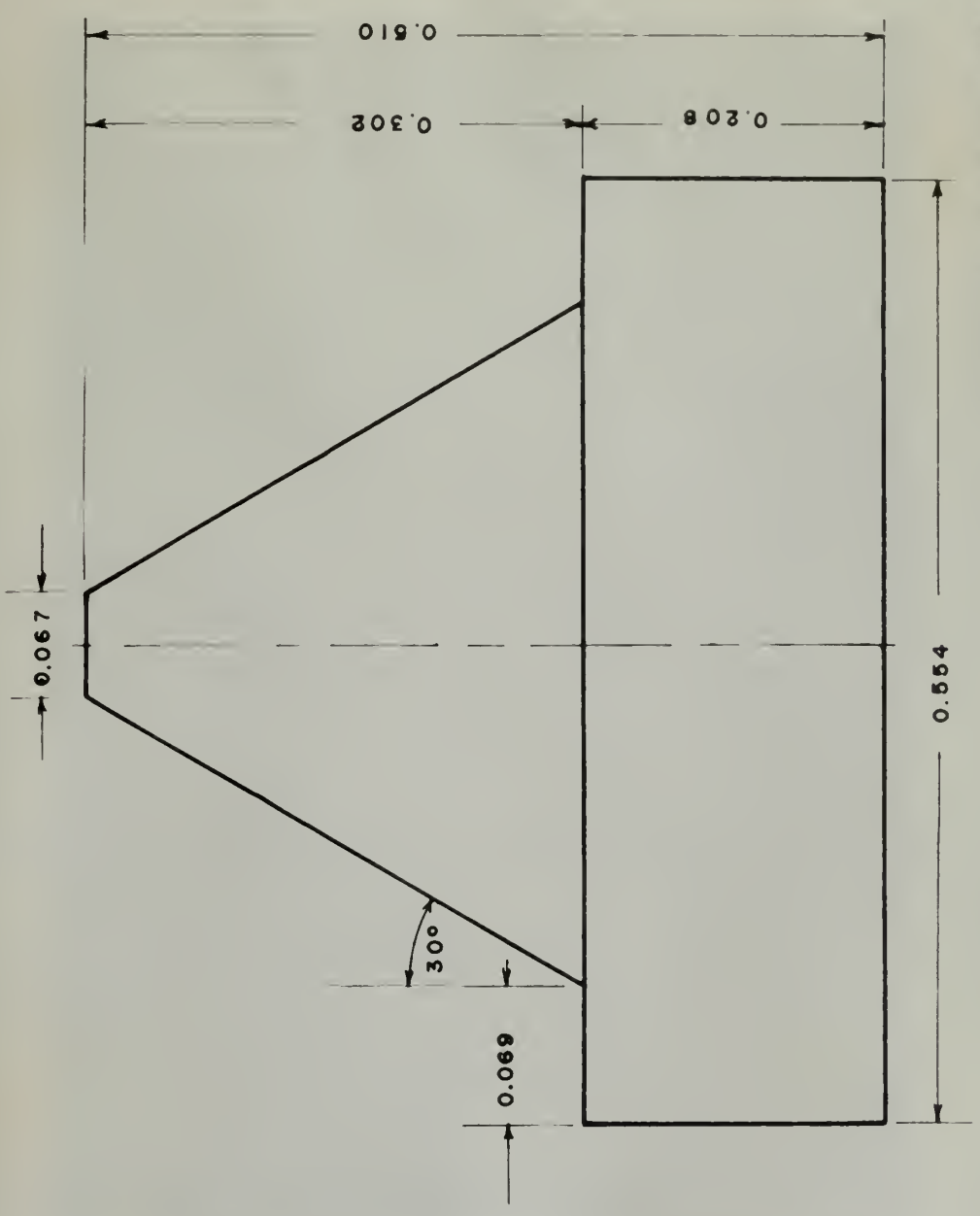
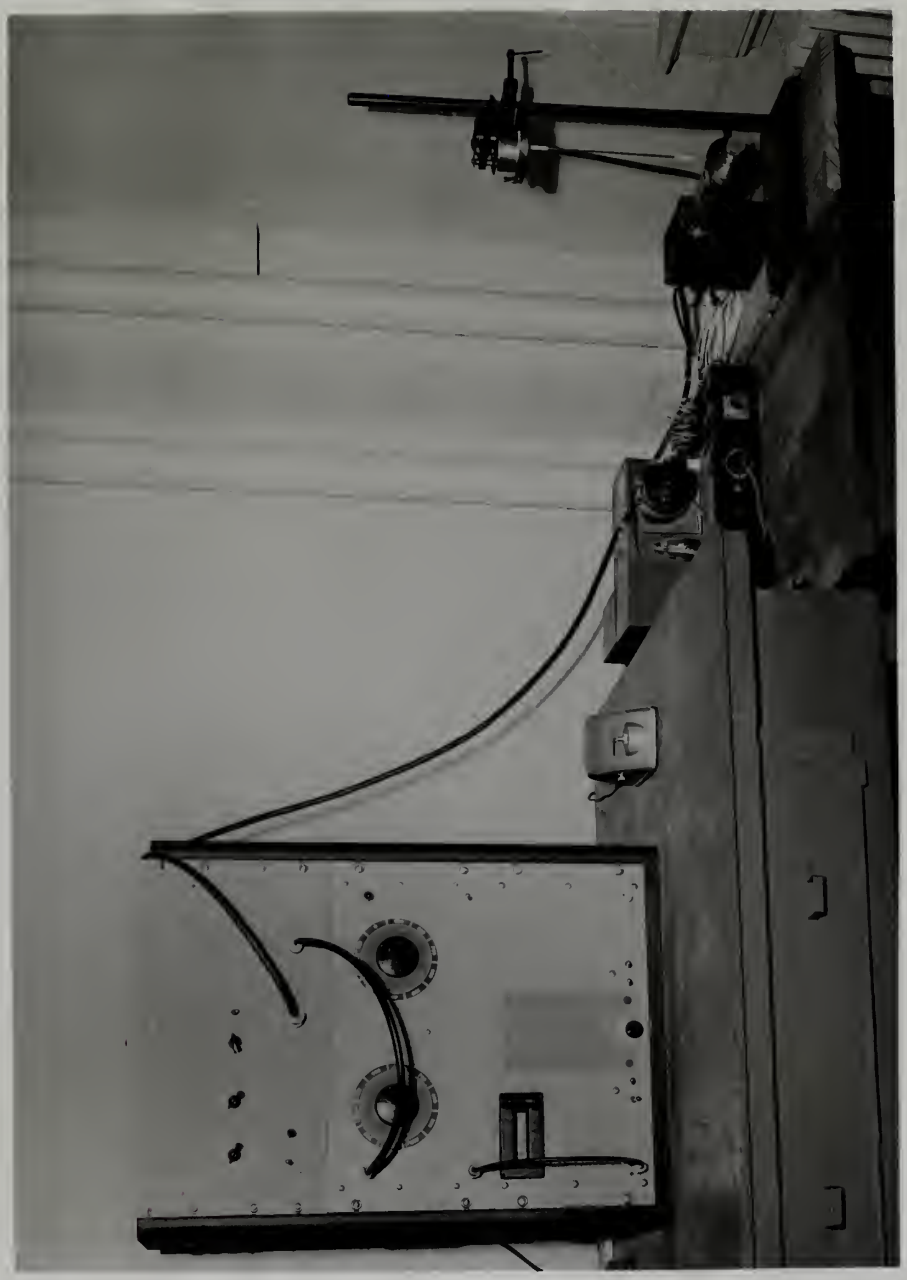


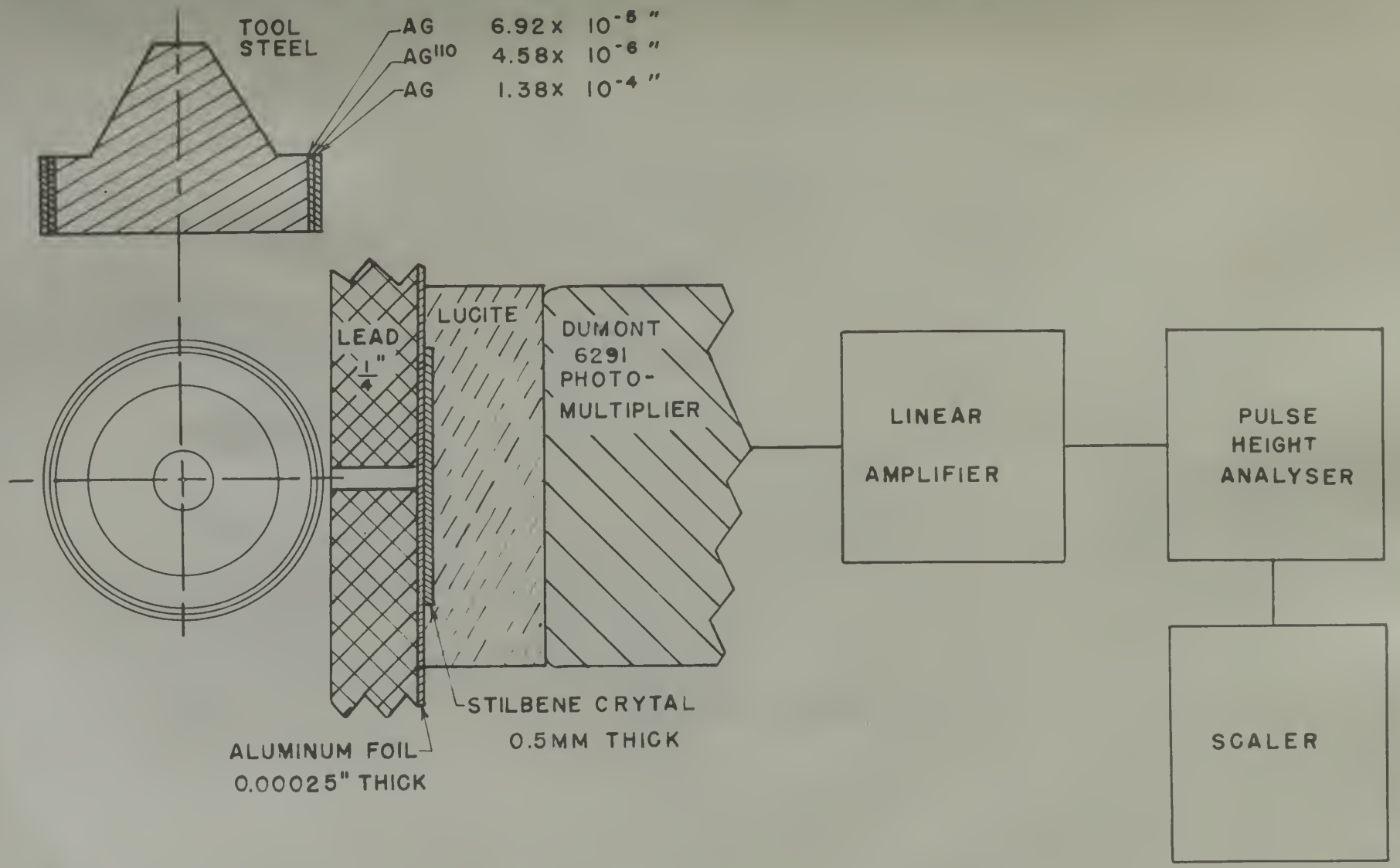
FIGURE 14

ROTOR DIMENSIONS



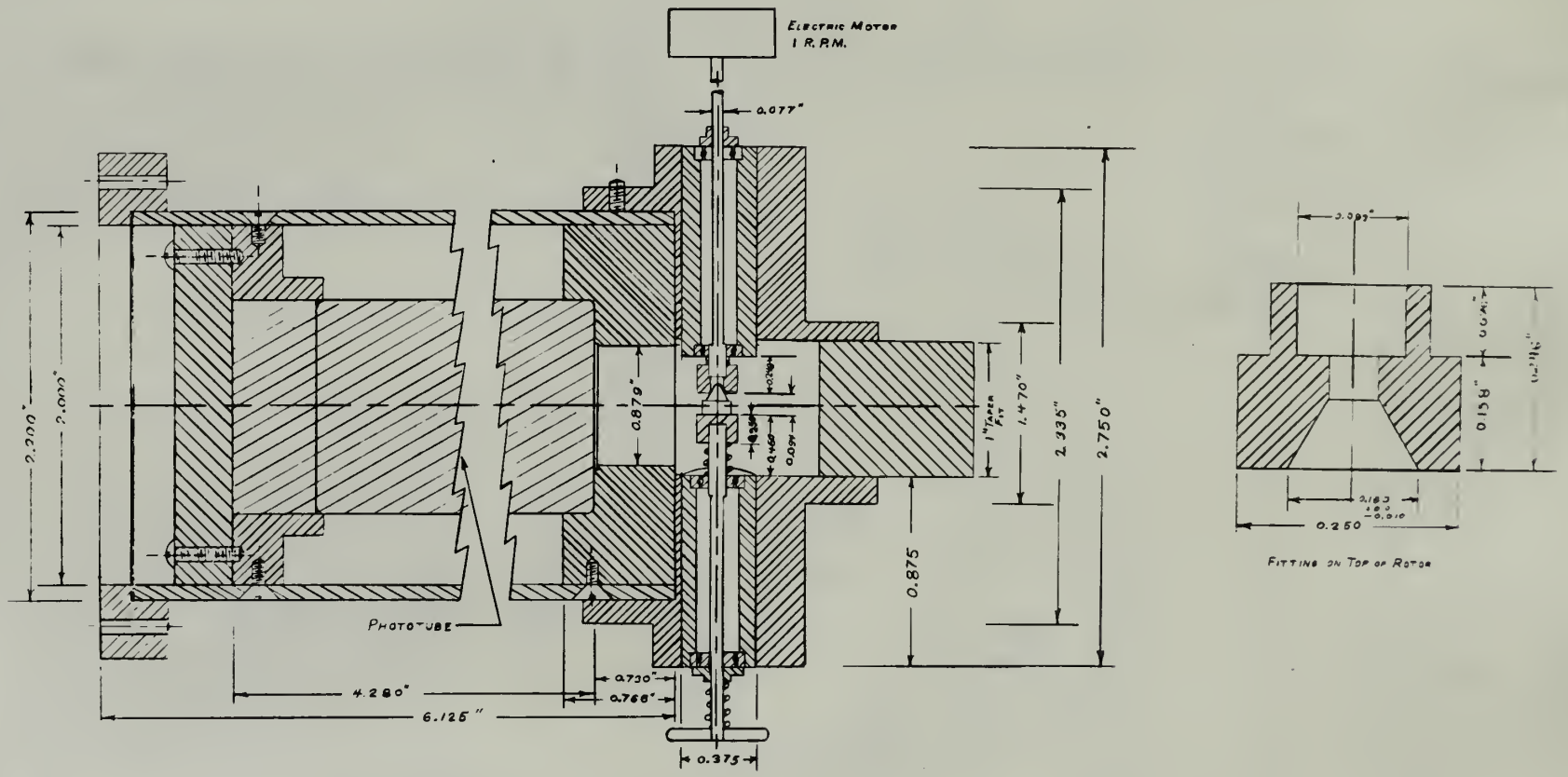
BETA-RAY DETECTION EQUIPMENT - FIGURE 15

FIGURE 15 - SECTION MICROSCOPIQUE DU CALCAIRE



ROTOR PREPARATION AND DETECTION EQUIPMENT FOR β-RAYS

FIGURE 16



Fitting on Top of Rotor

Phototube Scintilation Counter

Rouss Physical Laboratory

FIGURE 17

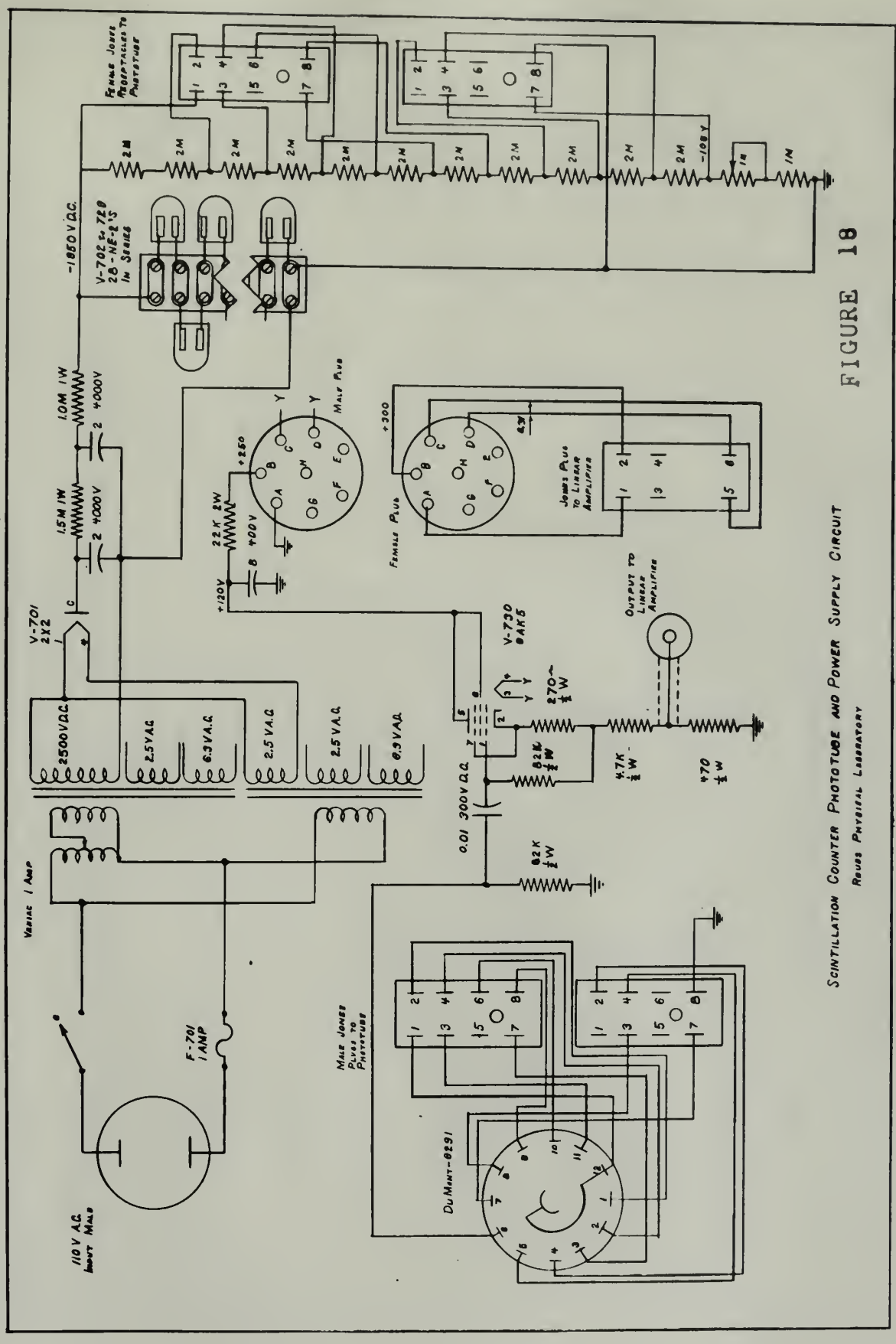


FIGURE 18

SCINTILLATION COUNTER PHOTOTUBE AND POWER SUPPLY CIRCUIT

REVES PHYSICAL LABORATORY

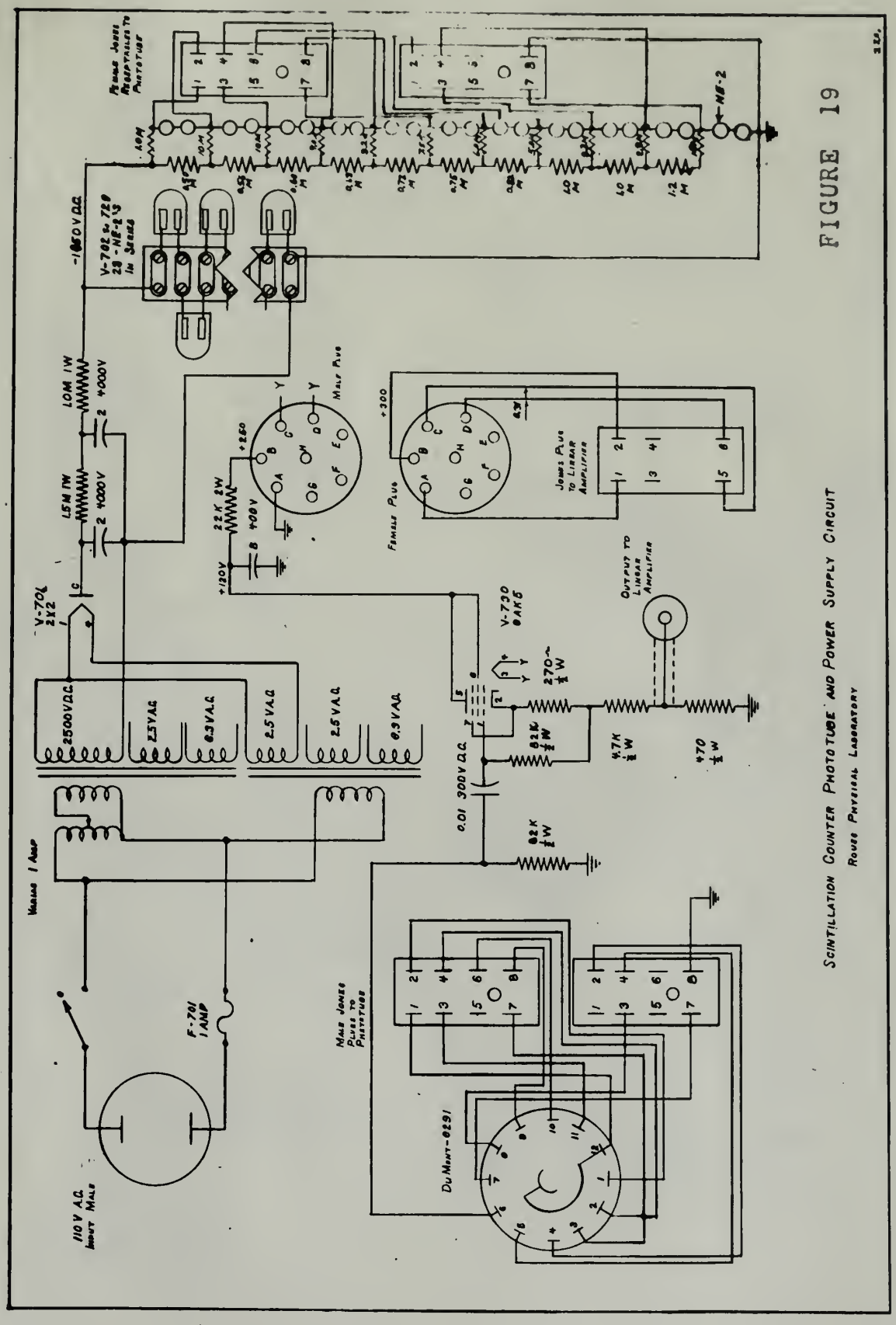


FIGURE 19

SCINTILLATION COUNTER PHOTOTUBE AND POWER SUPPLY CIRCUIT

ROUSE PHYSICAL LABORATORY

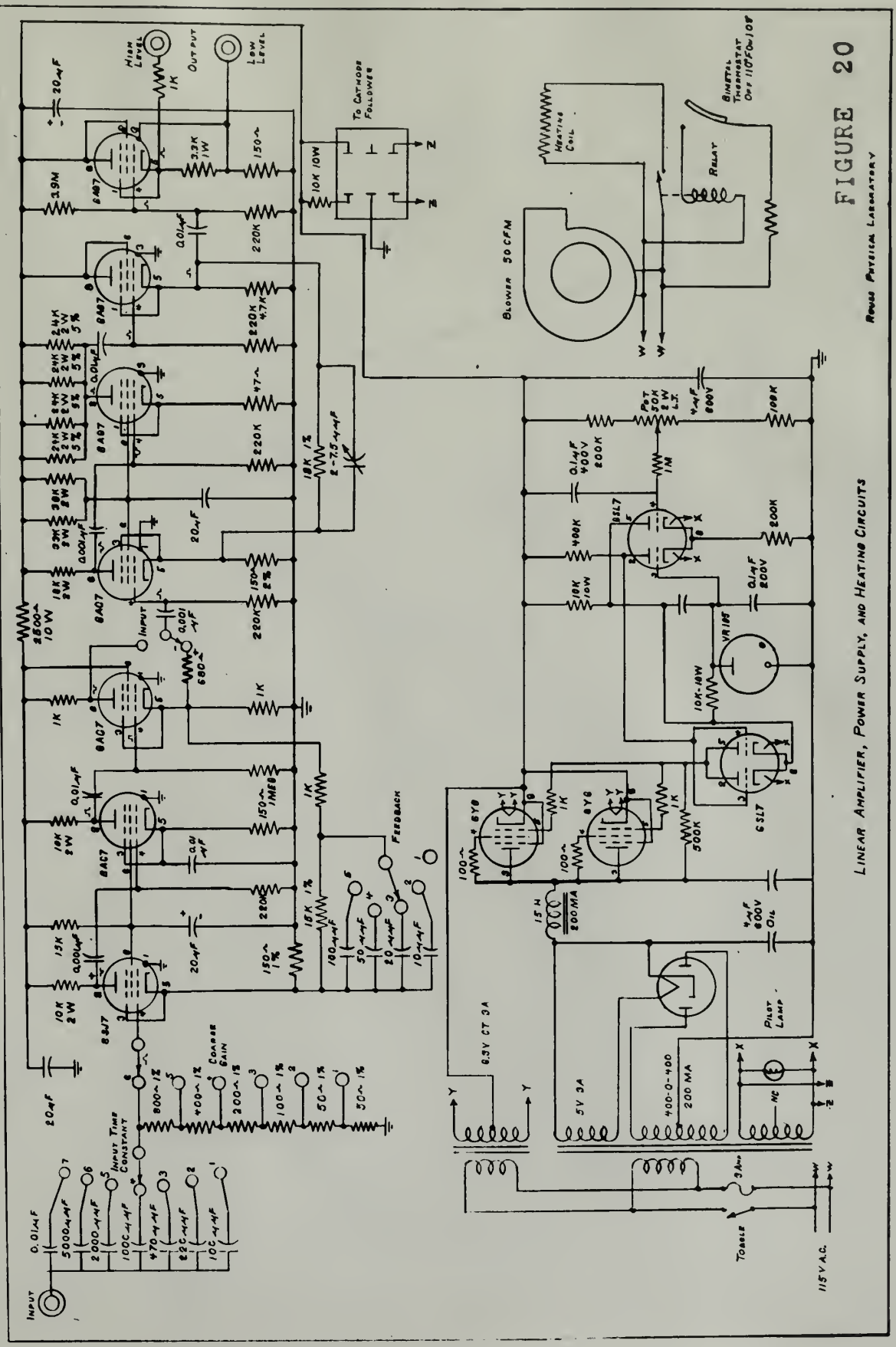
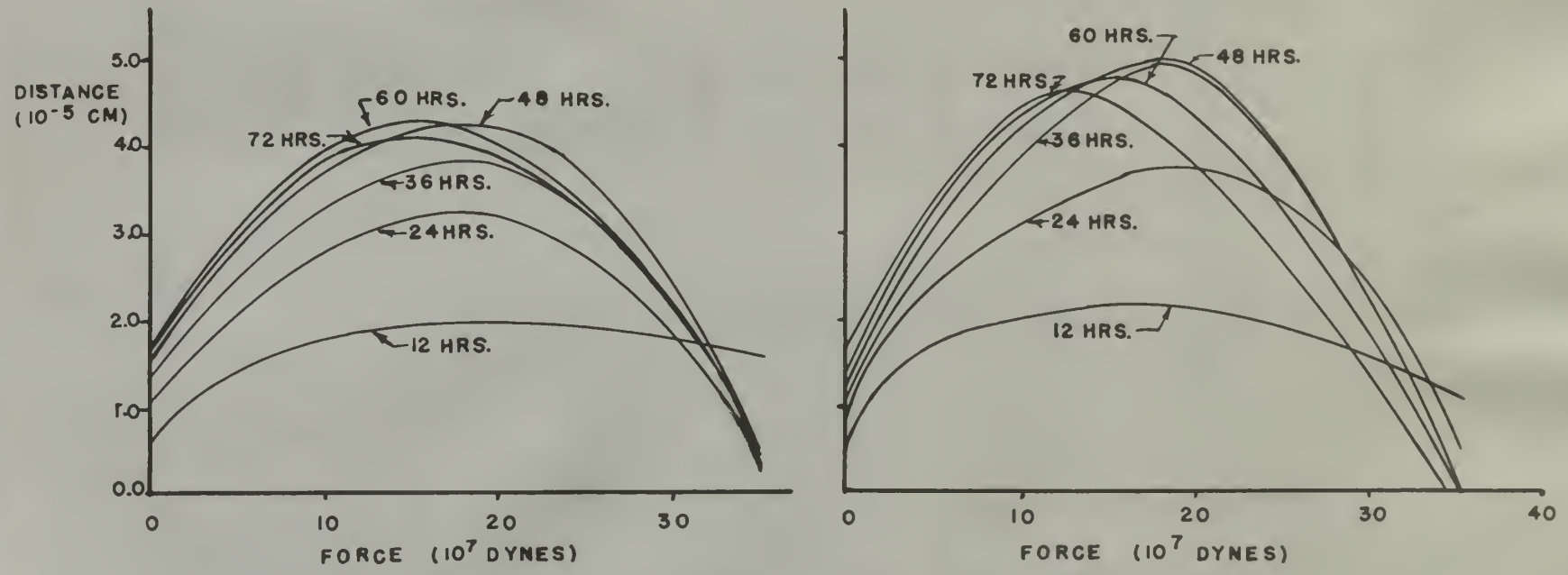


FIGURE 20

LINEAR AMPLIFIER, POWER SUPPLY, AND HEATING CIRCUITS

ROWE PHYSICAL LABORATORY



AVERAGE DISTANCE VS. APPLIED FORCE

FIGURE 21

APPENDIX I

Data for Typical Test Run.

Rotor velocity - 6,000 revolutions per second

Rotor temperature - 300 Degrees Centigrade

Time Hrs.	Control Rotor	Test Rotor	Adjusted Value	% Change
Rotor #21. Plated by the first method				
0	48,613	69,225	[illegible]	[illegible]
12	47,385	70,544	74,820	4.3
24	45,628	70,853	72,420	7.8
36	45,721	71,648	76,040	9.2
48	44,751	70,513	76,480	10.1
60	43,632	67,278	74,980	8.1
72	43,394	67,514	75,270	8.5
Rotor #26. Plated by the second method				
0	30,340	24,756		
12	28,954	24,216	25,760	4.1
24	28,365	24,466	26,190	6.2
36	28,094	24,723	26,730	8.2
48	28,043	24,783	26,800	8.3
60	29,531	26,195	26,920	9.1
72	29,613	25,892	26,580	7.5

APPENDIX I

Table 201. Index number of the gross domestic product

Year	Gross domestic product	Index number	Percentage change
1	46,013	89,353	
11	47,201	110,944	4.3
21	49,000	120,033	1.8
31	49,121	123,040	2.5
41	44,321	126,313	10.1
51	43,038	127,318	6.1
61	41,324	128,554	8.8

Table 202. Index of the economy output

4	39,345	84,124	
14	40,324	94,814	4.3
24	38,145	94,406	6.8
34	40,004	94,143	8.4
44	38,041	94,143	4.3
54	40,322	93,345	3.7
64	38,014	93,894	7.3

APPENDIX I

RESULTS

RPS	Percent Change in Counting Rate					
	12 Hrs.	24 Hrs.	36 Hrs.	48 Hrs.	60 Hrs.	72 Hrs.
First Plating Method						
0	0.5	1.4	1.6	2.3	2.6	3.1
2,000	3.3	3.9	4.1	3.9	4.7	5.1
4,000	4.2	6.2	5.8	9.6	6.8	9.7
6,000	4.3	7.8	9.2	10.1	8.1	8.5
8,000	3.2	-0.4	1.8	-1.3	1.1	-1.4
Second Plating Method						
0	1.3	2.2	2.8	3.7	3.1	2.0
2,000	1.1	3.5	3.2	2.9	3.2	3.4
4,000	3.5	4.8	5.9	7.3	7.3	---
6,000	4.1	6.2	8.2	8.3	9.1	7.5
8,000	3.3	1.0	0.5	0.9	1.1	1.3

ANNEXE 3

APPENDIX II

CALCULATIONS

I. Correction for temperature difference between test rotor and rotor mounted on the thermocouple.

A. Temperature difference as the result of heat conduction through thermocouple leads:

$$dQ/dt = -KA\,(d\theta/dx)$$

$d\theta = 192^{\circ}$

$dx = 7$ cm.

$A = (0.0375)^2$

$K = K_{iron} + K_{constantan} = 0.20 + 0.054 = 0.254$

$(dQ/dt)_{loss} = -(0.254)\,(\quad)\,(0.0375)^2(192/7) =$

-3.06×10^{-2} cal/sec.

Radiation with walls:

$dQ/dt = A\,\quad\,(\theta_w^4 - \theta^4)$

$\quad = 222^{\circ}$C. $= 495^{\circ}$ K.

$\theta_w = 50^{\circ}$ C. $= 323^{\circ}$ K.

$A = 0.9738$ cm^2

$\quad = 0.40$ (steel)

$\quad = 5.66 \times 10^{-5}$ ergs/cm^{2}deg^{4}sec

$(dQ/dt)_{walls} = (0.9738)\,(0.40)\,(5.66 \times 10^{-5})$

$(109 \times 10^8 - 600 \times 10^8)$

$= -1,082,000$ ergs/sec

$= -2.58 \times 10^2$ cal/sec

$$(dq/dt) = (dq/dt)_{loss} + (dq/dt)_{walls}$$

$$= -3.06 \times 10^{-2} - 2.58 \times 10^{-2} = -5.64 \times 10^{-2}$$

Therefore the corrected temperature, θ_c, is given by:

$$\theta_c^4 = -(dq/dt)(1/A) + \theta_w^4$$

$$= -\frac{5.64 \times 10^{-2}}{(5.66 \times 10^{-5})(0.40)(0.9738)(2.39 \times 10^{8})} + 109 \times 10^{8}$$

$$= 1136 \times 10^{8}$$

or $\theta_c = 575^{\circ}K. = 302^{\circ}C.$

E. Heat supplied to overcome air friction:

$$\frac{K.E.}{t} = (1/2)I\left(\omega^2/t - t_0\right)$$

Since $I_{rotor} = 1.586 \times 10^{-2}$ gm cm^2

$$\frac{K.E.}{t} = (1/2)(1.586 \times 10^{-2})(2.39 \times 10^{-8}) \times (4\pi^2)(4000^2 - 3900^2) / (1.17 \times 10^{3})$$

$$= 5.31 \times 10^{-6} \text{ cal/sec.}$$

This amount of heat must be added to the rotor by the sustaining drive. Assuming that the energy thus added to the rotor were completely effective this would raise the rotor's temperature only a fraction of a degree.

II. Sample Calculation for Average Distance Radioactive Atoms Move:

(1 + percent change) (No. of counts at overlay thickness of 1.38×10^{-4} inches) = No. of counts for reference to Figure 3.

Considering a six percent change in the counting rate:

= (1.05) (63,000) = 66,800 counts

In Figure 3 this corresponds to an overlay thickness of:

1.26×10^{-4} inches

the average movement is then $(1.38 - 1.26) \times 10^{-4}$ inches

$= 1.2 \times 10^{-5}$ inches $= 3.1 \times 10^{-5}$ cm.

XXI. Change in Time Dependent Solution of Concentration as A Result of Centrifugal Force at 6,000 Revolutions per Second:

$$Z_n(x) = 87.2 \frac{n \cos \lambda_n x + (5.1 \times 10^{-10})(6000)^2 \lambda_n^2 \sin \lambda_n x}{[\lambda_n^2 + (5.1 \times 10^{-10})^2 (6000)^4 (4\pi^2)^2]}$$

$$= 87.2 \frac{n \cos \lambda_n x + 8.64 \times 10^{-1} \lambda_n \sin \lambda_n x}{[\lambda_n^2 + 0.745]\ 1/2}$$

$$= \frac{n \pi}{9.26 \times 10^{-4}}$$

The smallest λ_n is when n = 1:

$\lambda_1 = 9.94 \times 10^3$

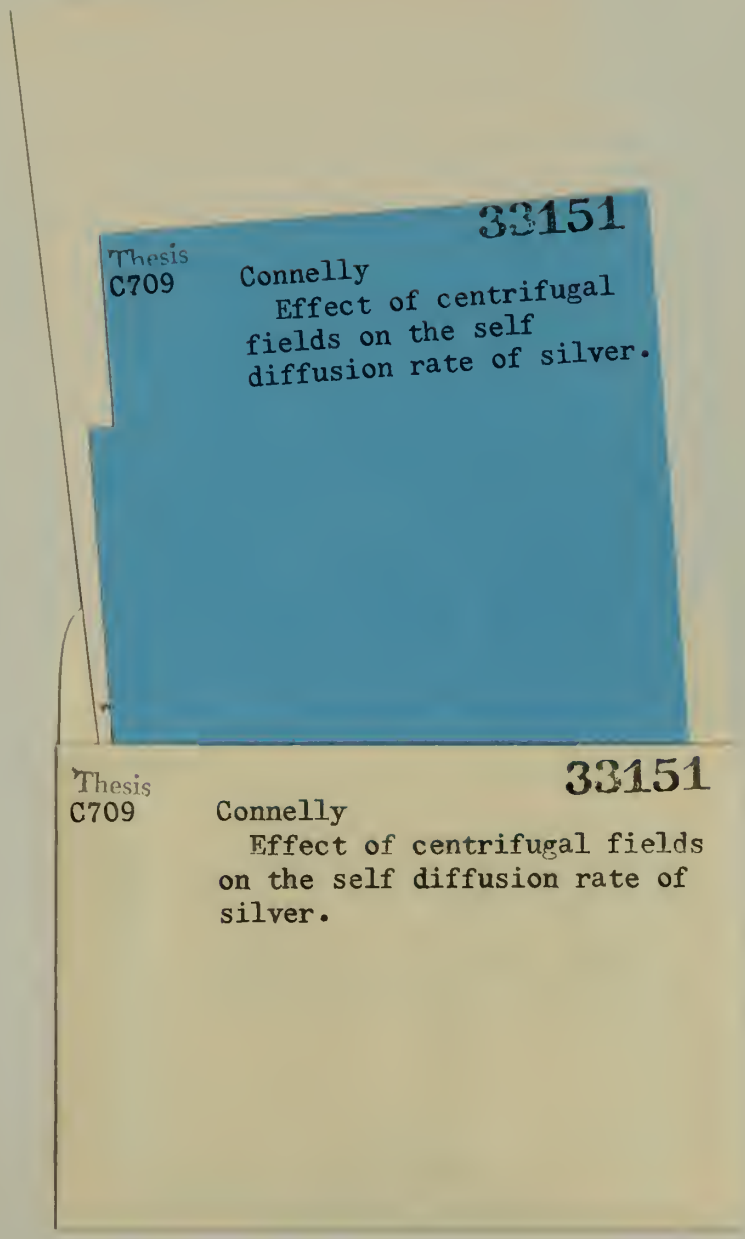
Then:

$$Z_n(x) = 87.2 \frac{5940 \cos \lambda_n x + 0.864 \lambda_n \sin \lambda_n x}{[(5940)^2 \lambda_n^2 + 0.745]}$$

$$= 87.2 (\cos \lambda_n x + 1.16 \times 10^{-4} \lambda_n \sin \lambda_n x)$$

When $\lambda_n = 0$:

$$Z_n(x) = 87.2 \cos \lambda_n x$$


33151
Thesis
C709
Connelly
Effect of centrifugal fields on the self diffusion rate of silver.

Thesis
C709
33151
Connelly
Effect of centrifugal fields on the self diffusion rate of silver.

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Effect of centrifugal fields on the self
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